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Published in Journal of Power Sources, 145, (2005), 683-690

Conceptual design and selection of a biodiesel fuel processor for a vehicle fuel cell APU

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Abstract

Within the European project BIOFEAT (Biodiesel Fuel Processor for a Fuel cell Auxiliary Power Unit for a Vehicle), a complete modular 10 kW_e biodiesel fuel processor capable of feeding a PEMFC will be developed, built and tested to generate electricity for a vehicle auxiliary power unit (APU). Tail pipe emissions reduction, increased use of renewable fuels, increase of hydrogen-fuel economy and efficient supply of present and future APU for road vehicles are the main project goals. Biodiesel is the chosen feedstock because it is a completely natural and thus renewable fuel. Three fuel processing options were taken into account at a conceptual design level and compared for hydrogen production: (i) autothermal reformer (ATR) with high and low temperature shift (HTS/LTS) reactors; (ii) autothermal reformer (ATR) with a single medium temperature shift (MTS) reactor; (iii) thermal cracker (TC) with high and low temperature shift (HTS/LTS) reactors. Based on a number of simulations (with the *AspenPlus*® software), the best operating conditions were determined (steam-to-carbon and O₂/C ratios, operating temperatures and pressures) for each process alternative. The selection of the preferential fuel processing option was consequently carried out, based on a number of criteria (efficiency, complexity, compactness, safety, controllability, emissions, ...): the ATR with both HTS and LTS reactors shows the most promising results, with a net electrical efficiency of 30% (LHV).

Keywords

Biodiesel, fuel processor, fuel cell, auxiliary power unit, conceptual design

Introduction

The finite nature of fossil energy sources, the strong European dependence on imported oil and the need for non-polluting energy conversion, all demand clean and efficient energy technologies that will have a vital role in the drive for sustainable development. Hydrogen, in combination with fuel cells, is likely to play a major role in future energy supply. The use of fuel cells, from small portable applications, through medium to large stationary power generation, to applications in the transportation sector, will offer in a medium to long term the prospect of significantly increased energy conversion efficiency coupled with little or even zero emissions of pollutants and greenhouse gases. Hydrogen could be a complementary energy carrier to electricity. Being intrinsically clean, it also permits the development of mechanisms for energy supply and demand management [1].

Although significant progress has been made in the development of fuel cells and hydrogen technologies, substantial barriers to successful commercialisation still remain. The costs of all fuel cells still need to be substantially reduced and their performance to be further improved in order to compete with other more established clean technologies [2]. Equally, there are major barriers to the implementation of hydrogen as an energy carrier: the development of production routes, a viable, safe and cost-effective method for its storage, the establishment of a hydrogen infrastructure and the public acceptance of hydrogen as a fuel. Research, technological development and demonstration have a crucial role to play in overcoming these barriers: therefore, the EU has supported these tasks in the area of hydrogen and fuel cell technology from the 1970s to the present days [3].

Concerning road transport, in the short-medium term on-board hydrogen production from hydrocarbon fuels has gained large importance for auxiliary power units (APU) based on fuel [4]. It is expected that the on-board power consumption in vehicles will play an increasingly important role: the on-board power demand has, in fact, increased significantly in the last decade and it is expected to grow even further. For passenger vehicles, Delphi Automotive Systems, an American vehicle electronic equipment supplier, predicts that the installed electric power on-board will increase to 10 kW_e before 2020 (Figure 1). This power will only be ensured by the auxiliary units in the vehicle and will not be used for traction purposes. If traction is taken into account as well, it is expected that the installed power will be twice this value [5].

Within such a context, the BIOFEAT project [6], involving beyond the authors other partners from the industry (Centro Ricerche FIAT, I; Johnson Matthey, UK; Bekaert, B; Scandiuzzi Advanced Technologies, I) and the academic world (Duisburg-Essen University, D), is expected to play an important role. The main purposes of the project are to increase fuel economy and to reduce tailpipe emissions by supplying the present and future auxiliary power requirements for peripheral systems in road vehicles (heating, ventilating and air conditioning system, on-board computers, steering by wire, brake by wire, in-car entertainment) using an auxiliary power unit (APU) incorporating a fuel cell fed by a biodiesel fuel processor. The specific objective of the BIOFEAT project is to design, build and demonstrate an on-board biodiesel fuel processor capable of feeding a fuel cell to generate electricity for the auxiliary power vehicle requirement for a family car or a truck. Provisional overall targets for the biodiesel fuel processor to be suitable for use in a production vehicle are listed in Table 1. The biodiesel fuel processor prototype being produced by the project will demonstrate as a minimum that these targets are achievable in production.

Biodiesel was selected because it is a 100% vegetable oil based and renewable fuel. Currently, biodiesel is produced mainly from field crops in Europe whereas elsewhere in the world, it is made from recycled cooking oil. In the past decade, biodiesel has been gaining worldwide popularity as an alternative energy source because of its many benefits. This environment friendly fuel reduces tailpipe emissions (it is practically sulphur-free, < 10 ppm), visible smoke and noxious odours compared to conventional diesel. Because biodiesel is non-toxic and biodegradable, handling and storage are safer than conventional petroleum diesel fuel. The cost compares well with other alternative fuels, despite it is still higher than those of oil distillates. Biodiesel can be used in existing engines and fuel injection equipment without a major performance penalty, it gives nearly the same km-per-litre performance and can be stored and dispensed in the same manner as normal diesel oil. Biodiesel has also a very high flash point making it one of the safest alternative fuels. Biodiesel is the only alternative fuel that produces basically no emissions during manufacture.

General process description and basis of design

A biodiesel fuel processor consists of a number of sub-units. A schematic flowsheet of a biodiesel fuel processor is shown in Figure 2. The main unit is the catalytic primary processor reactor for

hydrogen gross production: the two possible solutions considered in BIOFEAT are the autothermal reforming (ATR) and the thermal cracking (TC) of biodiesel [7].

After the primary step, secondary units for both the CO clean-up process and the simultaneous increase of the hydrogen flow rate are employed: the H₂ content from the reformat gas can be increased through the water gas shift (WGS) reaction by converting the CO with steam to CO₂ and H₂. The shift reaction itself is exothermic and therefore favoured at low temperatures. To limit the exit concentration of CO at around 2000-5000 ppm, normally a series of adiabatic reactors with intercooling is applied: the high thermal shift (HTS) reactor is operating at 300–350°C followed by a low thermal shift (LTS) reactor operating at 190–260°C [8]. It is also possible to use a single step conversion, using a mid thermal shift (MTS) reactor operating at 250–350°C.

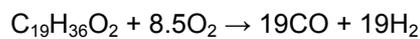
The product stream from the WGS reactors still contains CO because of equilibrium limitations. As the polymeric fuel cell (FC) is poisoned by this component, the maximum allowable CO concentration in the fed gas should be less than 50 ppm and preferably less than 10 ppm. An additional reactor of preferential oxidation (PROX) is thus required to completely remove the CO by oxidation to CO₂ on a noble metal catalyst. However, due to the presence of O₂, H₂ is oxidized as well as a side reaction. Air is normally used as the oxidizing agent. In this concept study, the PROX reaction is assumed to take place in an isothermal bed reactor at 150°C after the last shift reactor [9]. Due to the exothermic reaction, a reactor temperature control is necessary to maintain selectivity and limit side reactions such as reverse WGS and methanation.

The exothermic reaction in the FC is carried out at a constant temperature of about 80°C, which is maintained by cooling with demi-water. Normally, H₂ utilization is about 80%, leaving 20% of the H₂ unconverted in the anode outlet stream. Such anode side stream, containing H₂, is then burned in an afterburner, to recover the heat required to pre-heat the feed streams and to produce the required amount of steam for the fuel processor.

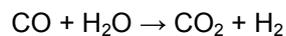
Another important issue of the whole fuel processor is water management. Water recovery from most of the product streams and recycling to the fuel processor optimize the fuel processor efficiency and minimizes the need for a large water storage vessel on-board. In the ideal case, no water make up has to be conceived since the required minimum amount is produced in the reactors during the start-up, enabling shorter transients. This entails that no water will freeze when the vehicle is not used. A series of auxiliary units for the balance of plant of the whole system

(afterburner for the combustion of hydrogen exhaust gas from the fuel cell, heat exchangers for the internal heat recoveries, water recovery radiators, air compressor, water and fuel pumps) are though necessary to proper operate the fuel processor with the fuel cell.

In the ATR process, the biodiesel is fed to the reactor together with steam and air where it is converted to H₂. This process is a combination of catalytic partial oxidation (CPOX) and steam reforming (SR). The endothermic heat of -the SR reaction is provided by the catalytic exothermal CPOX. Advantages are that no external fuel is required, flexibility in feed selection, higher turndown ratio and a fast start-up time with respect to steam reforming. The following reactions take place in the ATR; a part of the feed is oxidized partially as follows:



The remaining feed is catalytically reformed via the SR and WGS reactions:

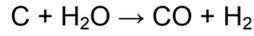


Additional side reactions, like carbon formation and complete oxidation reactions, are prevented by carefully selecting the operating conditions and properly mixing of the process inlet streams. The reforming reaction is carried out at an operating temperature of about 750°C.

The thermal cracker (TC) biodiesel fuel processor instead consists of a two-reactor system, in which one reactor, the cracker, is used for H₂ production, while the other one is being regenerated by gasification of the deposited solid C with steam yielding H₂, CO, CO₂ and CH₄ [10]. The regeneration may be carried out entirely by steam or by a combination of steam and air. The product gas streams of the TC and the gasification unit are combined, cooled down to the shift inlet temperature and then fed to the shift reactor and the CO-purification step. First, the cracking of biodiesel takes place for the production of H₂:



The regeneration process of the deposited C is then carried out in parallel, in a second reactor, via endothermic gasification:



The cracking and gasification reactions are both carried out at a operating temperature of 900°C.

Within the BIOFEAT project, three fuel processor configurations were taken into considerations, schematically represented in Figures 3, 4 and 5, respectively:

- autothermal reformer with high and low temperature shift reactors (case ATR_1),
- autothermal reformer with a single medium temperature shift reactor (case ATR_2).
- thermal cracker with high and low temperature shift reactors (case TC).

For the three fuel processor options, a series of systems simulations in *AspenPlus*® were carried out to determine the best operating conditions, in line with the main part of EU industrial project in the field [11]. The starting assumption for the basic design modelling of the different fuel processing systems for the conversion of biodiesel are listed in Table 2, whereas Table 3 illustrates the inlet conditions considered in the simulations for the different fuel processor options. The fuel processor (FP) and APU (or system) efficiency (η) are defined as follow [12]:

$$\eta_{FP} = \frac{m_{\text{H}_2,FP} \cdot \text{LHV}_{\text{H}_2}}{m_{bd} \cdot \text{LHV}_{bd}}$$

$$\eta_{APU} = \eta_{util} \cdot \eta_{FC} \cdot \eta_{FP} \cdot \eta_{aux}$$

where:

LHV_{H_2} , LHV_{bd} = lower heating values of hydrogen and biodiesel [J/mol];

$m_{\text{H}_2,FP}$, m_{bd} = molar flow rates of produced hydrogen and of biodiesel [mol/s];

η_{util} = fraction of hydrogen converted in the fuel cell (utilisation) [-];

η_{FC} = fuel cell efficiency [-];

η_{aux} = parasitic losses (pumps, compressors, inverters, etc.) $(1 - P_{aux}/P_{FC})$ [-]

Conceptual process design

The fuel processor options above-mentioned were evaluated taking into consideration the concentration values for the various components, the auxiliary power/heat demand, the water management and the system and the fuel processor efficiency. To select the most likely operating conditions for each option (operating pressure, steam-to-carbon and oxygen-to-carbon ratios, S/C

and O_2/C respectively), a series of simulations in *AspenPlus*® were performed. The selected results are shown in Table 4.

The major difference in H_2 concentration values between TC and ATR is due to the fact that the nitrogen in air dilutes the ATR streams and therefore results in a lower H_2 mole fraction. Conversely, the absolute H_2 amount (mol/s) is approximately similar for these two process options. This is not the case for CO: its presence is determined by the number of WGS reactors and their operating temperature. Two WGS reactors (operating at high and low temperature respectively), are beneficial for the CO concentration (ATR_1), as the operating temperature influences the equilibrium of the WGS reaction and the second WGS reactor can operate at very low temperatures. By using a single MTS reactor, higher CO outlet concentration (ATR_2) are obtained. Moreover, the higher the steam-to-carbon ratio, the lower the CO amount at the shift outlet: an increase in the S/C value in fact boosts the WGS reaction and minimises the chance of coke formation if the ATR reactor [12].

The choice of the operating pressure is a trade-off between the auxiliary power consumption (compressor) and the simplification in water balance achievement. At low pressures, additional heat is required for the fuel cell circuit, for which additional biodiesel has to be burnt. Elevated pressures imply higher power consumption by pumps and compressors. However, as the fuel cell requires a humidified airflow as a cathode feed stream, an increased operating pressure results in a higher partial pressure of water. Hence, the operability and sizing of the water management is favoured by higher operating pressures. Increasing the pressure reduces in fact the heat demand in the system and approximately at 1.3 bar, the system produces sufficient heat to recover its own demand. Above this pressure, the parasitic losses become more pronounced.

The fuel processor and the system efficiency are dependent on the H_2 amount produced by the fuel processor and the biodiesel amount required for the H_2 . The results for the system efficiency are shown in Figure 6. None of the simulated systems is able to meet the (net) electric system efficiency of 35% for the given assumptions (for the demonstration system (see table 2)). Even neglecting the parasitic losses of the pumps and air compressors, the net electric efficiency is approximately 29%. Interesting is that the effect of the steam-to-carbon ratio for the ATR_2 system is much higher than with ATR_1: this is caused by the ATR_2 system configuration. For a steam-to-carbon ratio of 2.5, the system resembles the ATR_1 one, where the process water for the

reactor is heated by using the cooling duty of the downstream process streams. At a steam-to-carbon ratio of 2, this same cooling duty is too large for the heating duty required for steam production. The cooling of the MTS outlet stream is therefore carried out by the humidification water stream for the fuel cell cathode. The efficiency can be increased by selecting another operating voltage of the fuel cell. However, a higher operating voltage also means higher costs due to a larger stack. For the conceptual design, the operating voltage has been set on 0.6V and is kept constant for each of the simulated cases. The target minimum fuel processor efficiency of 75% is only obtained for all the ATR_1 systems and for the TC systems with operating pressure larger than approximately 1.3 bara. The ATR_2 systems cannot meet this requirement at any condition, mainly because a relative large amount of hydrogen is burned in the CO-PROX reactor. Hydrogen production might be increased by a more intensive quenching before the MTS reactor. However, the injection of more water lowers the inlet temperature of the shift reactor. This means that the MTS outlet temperature is also lower and that less heat can be exchanged with the water stream required for the fuel cell humidification. Additional biodiesel should be burned in that case to provide the extra heat, thereby limiting the net electrical efficiency.

Fuel processor selection

The selection was based on a number of selection criteria among which water management easiness, fuel processor and system efficiency, system controllability, dynamic behaviour, total system volume and emissions. Some of these criteria may affect one another: for instance a high system efficiency (and a high life time) can be achieved by a small loading related to the fuel cell area, according to a small current density; on the other hand, this leads to larger volume, weight and rising costs.

The highest system efficiency found for the three systems is reached by the TC option, but it is only slightly higher than ATR_1 option. The least attractive option from this viewpoint is ATR_2.

As for the water management, it is of utmost importance that a sufficiently large driving force exist with the ambient temperature to cool down the process streams and recover water by condensation; none of the considered systems are critical. For the TC option, the cooling temperature difference is the highest and is therefore selected as preferential option.

In transportation, space is a limiting factor and the system volume should be minimized as much as possible. Moreover, smaller systems are easier to control (e.g. temperature control) than larger systems. The type of catalyst used, largely determines the volume of the reactor, active catalyst and short contact times are required to achieve a low reactor volume. Figure 7 shows the overall catalyst volumes related to the three different fuel processing options, calculated on the grounds of the results of an extensive catalyst testing campaign carried out within the BIOFEAT project. The smallest system resulted to be the TC one, followed by the ATR₁ and the ATR₂.

ATR reactors are though very easy to control. The reactions are taking place in a single reactor and can be controlled by adjusting the process streams (air, steam or biodiesel). The controllability of the TC reactor is indeed more difficult, due to the presence of the two reactors, one for operation, the other for regeneration.

At this stage it is impossible to make a complete detailed cost calculation for the various systems. Therefore, a more quantitative approach is used for the comparison of the systems based on the relative cost, determined in comparison to the expected number of process units present in the fuel processing system (number of reactors, heat exchangers, pumps and compressors., water condensers).

Owing the absence of one shift reactor in the ATR₂ option, the fuel processor requires one reactor and one heat exchanger less than ATR₁. However, the additional bed that is required in the PROX reactor, because of the higher CO-concentration, compensates this advantage. Especially in the TC option, three heat exchangers are required to cool and heat three process streams from more than 900°C. These exchangers require expensive materials of construction and are therefore expensive. Moreover, the regeneration process to remove the deposited carbon from the catalyst is difficult and it is likely that not all the carbon is removed in the available regeneration time. Since this affects the catalyst activity and lifetime, replacement of the catalyst will be more frequent than for the ATR fuel processors. Consequently, the cost will also be higher: both ATR options are thus favoured.

The emissions formed in the processes are ideally CO₂ and H₂O. However, due to the high temperatures in the process other components can be formed as well, like NO_x and particulate matter, especially for the TC option. Another source of emissions is the afterburner burning the anode off gas. From that point of view, the TC option is less attractive than both ATR options.

For transportation purposes, in order to make the reformer as dynamic as possible, the system should be constructed in a compact way and light-weight, which enables short residence times of the gases in the different sections and short times for the temperature to reach its new steady state. Among the three selected options, the ATR_1 seems easier to control than the ATR_2 and the TC, respectively.

With all the considered factors, a ranking was given for each of the systems. However, the mentioned selection criteria cannot be rated equally because some will be more important than others. For example, a high system efficiency is desirable, but if that results in lower controllability or higher cost, the lower efficiency will be favoured. Cost and controllability are generally more important criteria in the system selection procedure. This is also the case for the dynamic behaviour of the system. For that reason, weighing factors have been applied for the mentioned criteria. For each of the criteria the matter of importance is weighed at a scale from 1 to 5. Here, the factor 5 means "very important", while the factor 1 means "not important". The results are shown in Table 5: summing up all the factors the autothermal reformer with two shift reactors (ATR_1) has the highest ranking and seems therefore the most promising fuel processor option.

Conclusions

A fuel processor will be developed, build and tested within the European project BIOFEAT. The purpose is to provide electricity for the APU in future vehicles. In this study, three fuel processor options were compared for H₂ production from biodiesel. Based on a number of conceptual design simulations, the most likely process conditions were determined. The selection of the preferential fuel processor option was consequently carried out. As a result, the ATR_1 fuel processor (autothermal FP with HT and LT WGS steps) was found to be the most promising to be integrated in the APU unit. This unit will operate at a slight overpressure (1.3 bar) and with a steam-to-carbon ratio of 2.5. The overall APU system efficiency of the ATR_1 option is expected to be approximately 29%.

Acknowledgments

Funding of the European Union is gratefully acknowledged (EU project BIOFEAT nr ENK-CT-2002-00612: Biodiesel fuel processor for a fuel cell auxiliary power unit for a vehicle). A special thank to

all the involve partners for their fruitful co-operation (Centro Ricerche FIAT, Duisburg-Essen University, Scandiuzzi Advanced Technologies, Bekaert, Johnson Matthey).

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Tables captions

Table 1 – Provisional overall BIOFEAT targets for the biodiesel fuel processor at a production vehicle level.

Table 2 – Starting points for the design of the biodiesel fuel processor demonstration unit.

Table 3 – Inlet conditions considered in the simulations for the different fuel processor options.

Table 4 – Simulation results for the best selected operating conditions for the three fuel processor options.

Table 5 – Fuel processing option ranking: total overview.

Figures captions

Figure 1 – Development of the on-board power demand for passenger vehicles (courtesy of Delphi automotive systems).

Figure 2 – Schematic flowsheet of a biodiesel fuel processor.

Figure 3 - Schematic flowsheet of the ATR_1 fuel processing option

Figure 4 - Schematic flowsheet of the ATR_2 fuel processing option

Figure 5 - Schematic flowsheet of the TC fuel processing option

Figure 6 – Total system efficiency as function of the operating pressure.

Figure 7 – Total catalyst volumes for the various fuel processor options

Tables

Table 1

1.	size ≤ 0.75 l/kW _e compatible with the available on board space
2.	specific weight: ≤ 1.5 kg/kW _e
3.	maximum start-up time: 1 min. to full-power
4.	dynamic response: ≤ 5 sec (to sudden variations in the auxiliary power requirement)
5.	reliability > 10.000 h or 120.000 km
6.	efficiency of biodiesel-to-hydrogen conversion: $> 85\%$
7.	overall efficiency of chemical-to-electric energy: $> 45\%$
8.	cost < 10 €/kW _e (industrial mass production)
9.	CO content in H ₂ -rich stream: < 50 ppm
10.	unburned residues of biodiesel (HC) in the exhaust: < 5 ppm
11.	NO _x content in the exhaust gases: negligible

Table 2

Process parameters	Value unit demonstration unit
Biodiesel fuel supply: - inlet temperature	25°C
Primary processor reactor, outer temperature range: - autothermal reformer - thermal cracker	700-750°C 900-930°C
Fuel spec to fuel cell: - H ₂ flow - CO concentration - Sulphur concentration - Temperature - Pressure - Side reactions, current leakage - Oxygen utilisation - Cell voltage - Humidification constant	0.2 kg/h < 10 ppm < 1 ppm 80°C < 3 bar 2% 50% 0.6 V 80-100%
Fuel cell: - H ₂ utilisation	80%
Fuel processor: - Water inlet temperature - Ambient temperature (min/max)	25°C -10 – 40°C
Reactors: - Heat loss - PROX reactor selectivity (pessimistic value) - Thermal equilibrium in ATR, HT/MT/LT WGS	0 W 20% CO
Other process units: - Compressor efficiency - Pump efficiency - Minimum temperature difference heat exchangers	72% 65% 15°C
Afterburner: - Max outer temperature ATR - Max outer temperature TC	< 650°C < 1500°C
Power produced/demand: - Power produced - Auxiliary power	10 kW _e < 2 kW _e
Efficiency (lower heating value, LHV): - System efficiency (DC) - Fuel processor efficiency	> 35% > 75%

Table 3

Fuel processor options	Reactor inlet temperatures	Pressure range	S/C range	O₂/C range
Case ATR_1: - ATR - HTS - LTS - PROX	325°C 398°C 240°C 150°C	1-3 bar	2-2.5	0.39-0.41
Case ATR_2: - ATR - MTS - PROX	325°C 300°C 150°C	1-3 bar	2-2.5	0.39-0.41
Case TC: - Cracker/regenerator - HTS - LTS - PROX	900°C 400°C 200°C 150°C	1-3 bar	2.5-3	-

Table 4

Fuel processor options	ATR_1	ATR_2	TC
Operating pressure [bar]	1.3	1.3	1.3
S/C ratio	2.5	2.5	3.0
O ₂ /C ratio	0.4	0.4	-
Fuel-cell inlet gas composition:			
- H ₂ [%]	29.07	25.46	47.33
- H ₂ O [%]	31.78	33.54	26.89
- CO ₂ [%]	14.81	14.01	18.15
- CH ₄ [%]	0.02	0.05	0.33
- N ₂ [%]	23.88	26.63	7.29
- CO [ppm]	< 10	< 10	< 10

Table 5

Selection criteria	Unweighted scores			Weighting factor	Weighted scores		
	ATR_1	ATR_2	TC		ATR_1	ATR_2	TC
System efficiency	2	1	3	4	8	4	12
Water management easiness	1	1	2	4	4	4	8
System volume	2	1	3	4	8	4	12
Controllability	3	3	1	5	15	15	5
Cost	2	2	1	5	10	10	5
Emissions	3	3	1	3	9	9	3
Dynamic behaviour/Operability	3	2	1	4	12	8	4
Total					66	54	49

“1” = low; “2” = fair; “3” = good

Figure 1

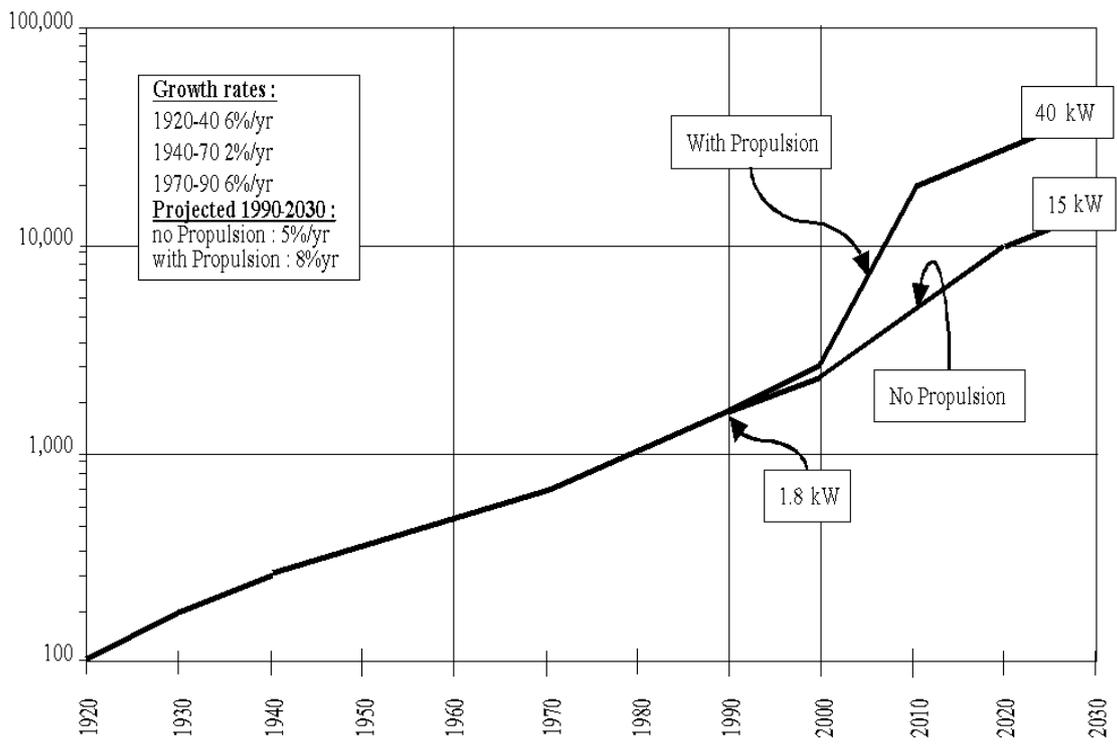


Figure 2

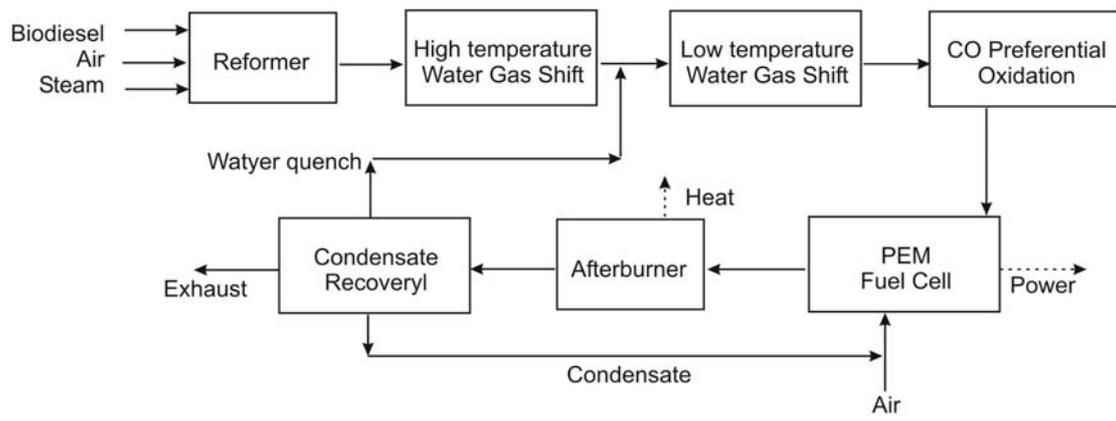


Figure 4

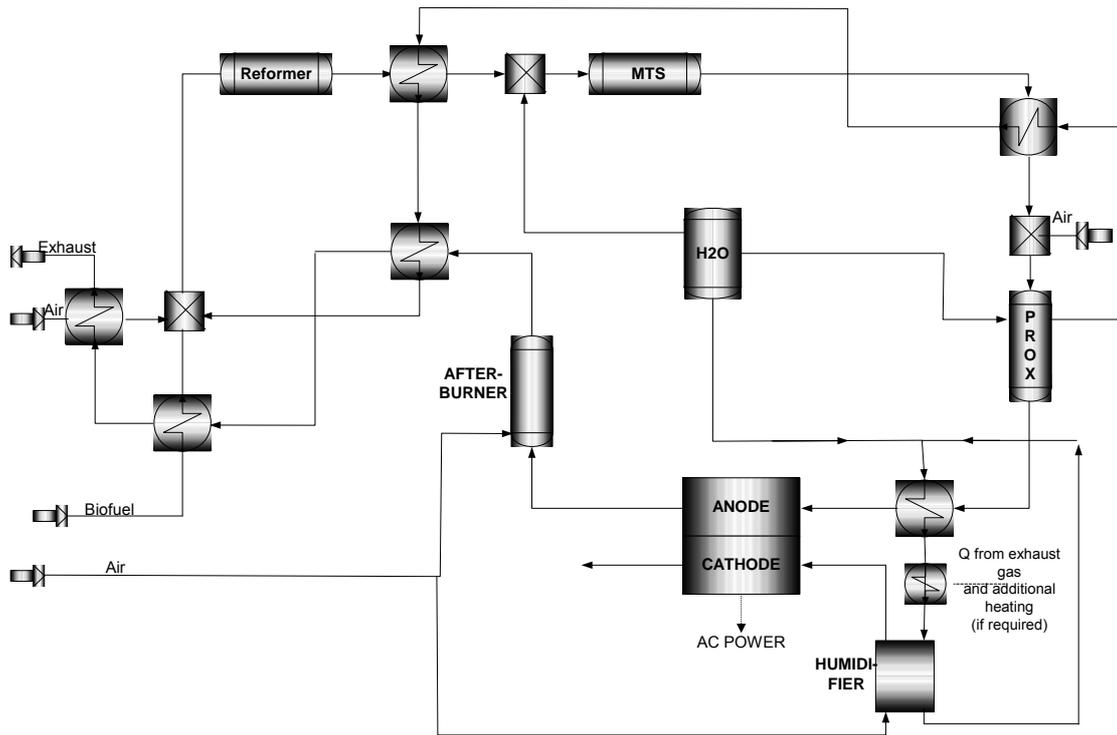


Figure 5

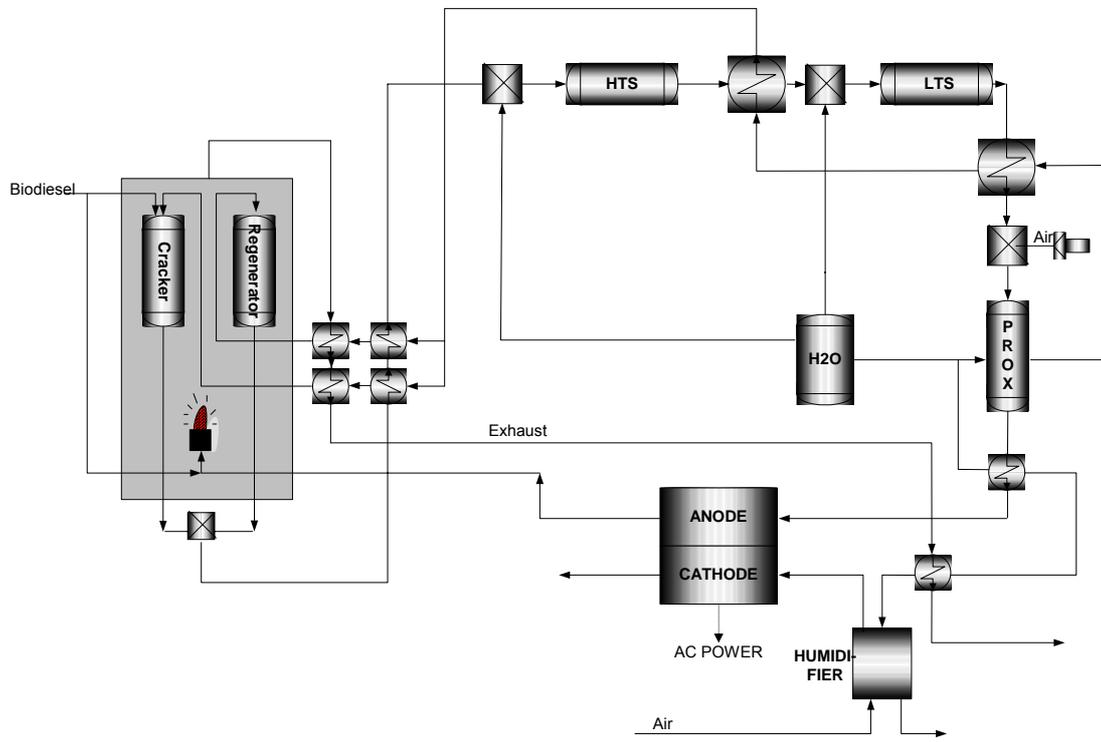


Figure 6

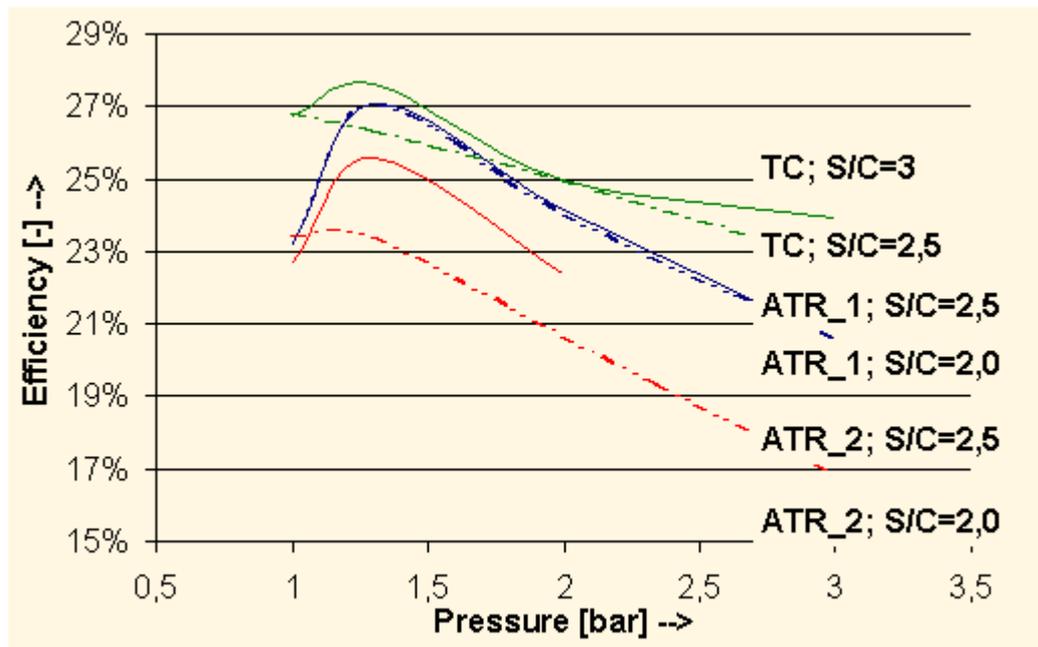


Figure 7

