

# BIOFEAT: Biodiesel fuel processor for a vehicle fuel cell auxiliary power unit<sup>☆</sup>

## Study of the feed system

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Received 30 November 2004; accepted 26 December 2004

Available online 31 May 2005

### Abstract

An integrated auxiliary power unit (APU) based on a 10 kW<sub>e</sub> integrated biodiesel fuel processor has been designed and is being developed. *Auto-thermal reforming* (ATR) and *thermal cracking* (TC) were considered for converting the fuel into a hydrogen-rich gas suitable for PEM fuel cells. The fuel processor includes also a gas clean-up system that will reduce the carbon monoxide in the primary processor exit gas to below 10 ppm via a new heat-integrated CO clean-up unit, based on the assembly of catalytic heat exchange plates, so as to meet the operational requirements of a PEMFC stack. This article is devoted to the study and selection of the proper feed strategy for the primary fuel processor. Different pre-treatment and feed alternatives (e.g. based on nozzles or simple coils) were devised and tested for the ATR processors, which turned out to be the preferred primary processing route. A nozzle-based strategy was finally selected along with special recommendations about the constituent materials and the operating procedures to be adopted to avoid coking and nozzle corrosion as well as to allow a wide turn down ratio.

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**Keywords:** Biodiesel; Fuel processor; Auxiliary power unit; Auto-thermal reforming; Thermal cracking; Vehicle

### 1. Introduction

The goal of the European-funded project *Biodiesel fuel processor for a fuel cell auxiliary power unit for a vehicle* (BIOFEAT) is to develop an auxiliary power unit (APU) based on a 10 kW<sub>e</sub> fuel cell stack fed by an integrated biodiesel fuel processor. The purposes of the project are the reduction of tailpipe emissions, the promotion of the use of renewable fuels and an increase in fuel economy compared to currently employed auxiliary power generation systems. The modular 10 kW<sub>e</sub> biodiesel fuel processor is capable of feeding a solid oxide or a polymeric membrane fuel cell stack

(PEMFC) that will generate electricity for the auxiliary power unit on a family car or a truck [1].

Significant market penetration of fuel cell systems for traction purposes is expected to take place only in long term. Higher application opportunities in the medium term (5–7 years) are expected in the field of auxiliary power generation for vehicles, thereby allowing the de-coupling of traction and peripherals-powering (air-conditioning, steering-by-wire, lights, . . .) needs [2]. These expectations are based on the lower nominal power of APUs and by the increasingly stable power demand required by the peripherals [3].

Biodiesel is the chosen feedstock in BIOFEAT, because it is a completely natural and renewable fuel. It is a 100% vegetable oil produced mainly from field crops in Europe, whereas elsewhere in the world, it is even made from recycled cooking oil. In the past decade, biodiesel has been gaining worldwide popularity as an alternative energy source

<sup>☆</sup> This paper was presented at the 2004 Fuel Cell Seminar, San Antonio, TX, USA.

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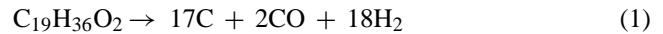
because of its many benefits. Besides the huge reduction in greenhouse gas emissions it entails, this environment friendly fuel reduces tailpipe emissions, visible smoke and noxious odours. Biodiesel is non-toxic and biodegradable, handling and storage are safer than conventional petroleum diesel fuel. Its cost compares well with other alternative fuels. Biodiesel also operates well in a conventional diesel engine with few engine modifications and no performance penalty.

## 2. Overall fuel processor description

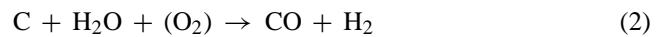
The BIOFEAT fuel processor consists of a number of stages (Fig. 1). The main component is the *primary fuel processor* that converts the feedstock into a hydrogen-rich gas. One of the major objectives of the project is to select either *thermal cracking* (TC) or *auto-thermal reforming* (ATR) as the preferred primary processing technique. Two separate research groups undertook this work (Johnson–Matthey

for the ATR, University of Duisburg-Essen for the TC). In parallel, two other activities considered the *requirements for conditioning the biodiesel* prior to entry into the primary fuel processor and the *treatment of the exit gas* from the primary fuel processor for a PEMFC system. The outlet gases of the primary fuel processor are indeed suitable for a SOFC, but a PEMFC requires a *gas clean-up system* to reduce the carbon monoxide level below 10 ppm.

The *thermal catalytic cracking* (TC) of biodiesel is an innovative approach for the generation of a hydrogen-rich gas with a gas quality that is similar, if not even better in terms of hydrogen concentration, to steam reforming. First, the fuel is decomposed to produce hydrogen:



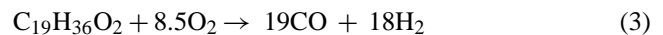
then, the solid carbon is regenerated via endothermic gasification:



The proposed biodiesel thermal cracker reactor consists of two reactors; one is used for hydrogen production by cracking; the other is being regenerated by gasification of the solid carbon with steam and air-yielding hydrogen, carbon monoxide, carbon dioxide and methane. The overall product gas contains about 70% hydrogen. The anodic off-gas from the fuel cell is fed back to the diesel burner of the cracker, with an increase in the overall system efficiency and a reduction of  $\text{NO}_x$ ,  $\text{SO}_2$  and soot emissions [4].

The *Auto-thermal reforming* (ATR) has been used to generate a hydrogen-rich reformate from a wide range of fuels including methanol, natural gas, LPG and more recently gasoline [5]. This process uses a combination of partial oxidation and steam reforming within the same catalyst bed to reform the fuel. Steam and air are fed into the reformer at a temperature ranging between 350 and 500 °C depending on the catalyst activity. The reactions that take place in an ATR reactor are reported below.

Partial oxidation:



Steam reforming and water gas-shift reactions:



After a preliminary study of both fuel-processing methods, the *auto-thermal reformer was selected as the best choice for the biodiesel processing*. The selection criteria for the most proper reforming strategy are described elsewhere [6,7].

In a biodiesel fuel-processing system based on auto-thermal reforming, typical reformate gases contain about

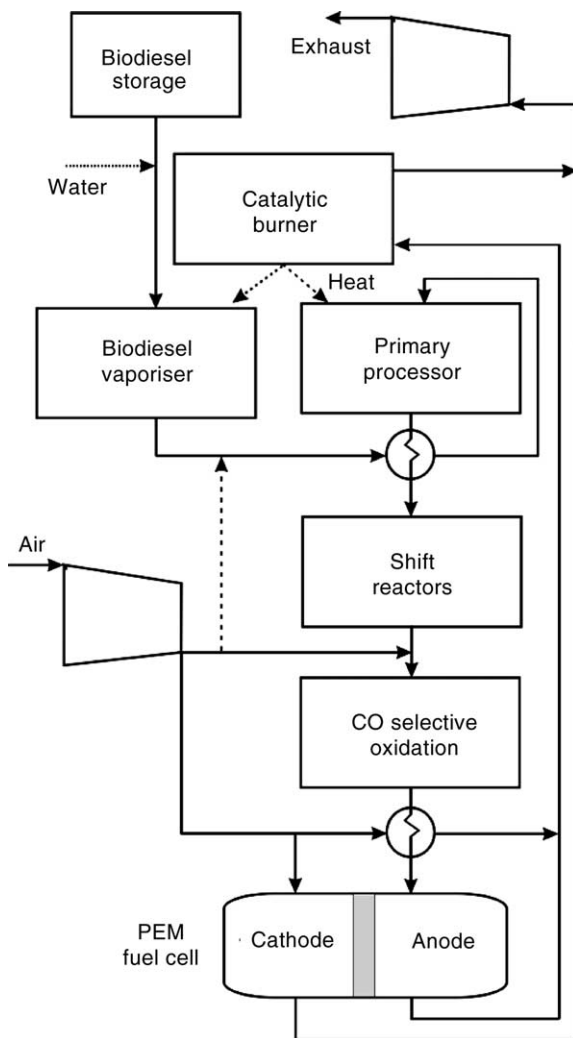


Fig. 1. System layout for the BIOFEAT APU based on the biodiesel fuel processor. After conversion in the primary reactor, the reformate stream is purified from CO in the water gas shift and in the CO clean-up reactors.

3–8 vol.% carbon monoxide at the outlet of the primary processor. After the primary biodiesel processing step, the hot synthesis gas has to be cooled down and further processing often takes place in two distinct water gas-shift reactors, where the carbon monoxide is converted with a water steam to carbon dioxide and additional hydrogen is produced due to the exothermic water-shift reaction (WGS):



A first high temperature shift (HTS) reactor, operating in the temperature range of 360–450 °C, allows high reaction rates, while a low temperature shift (LTS) converter, operating at about 210–270 °C, allows for a higher conversion. Noble metal (Pd, Pt)-based catalysts have proved to be active for the water gas shift as a replacement of Cu-Zn catalysts [8].

After this low-temperature shift reactor, the CO concentration is in the range of 0.3–1 vol.%; so, the CO has to be reduced further from the synthesis gas either by preferential oxidation (PROX or SELOX) or by selective methanation. On the other hand, mobile APU applications impose different requirements in many aspects. Any catalytic reactor for such appliances has to be cost-efficient, light, resistant to road vibrations (which would cause settlement of packed bed reactors) and capable of rapid and recurring start-up and shutdown. The use of tailored structures resembling those employed in industrial *corrugated plate heat exchangers* is a key feature of the project. Corrugated aluminium or stainless steel plates will allow good heat exchange between a Pt-Ru-catalysed CO PROX reaction, carried out on one side of the plates, and an air flow to be fed to the primary processor on the opposite side. Furthermore, such a CO PROX reactor configuration provides a low-pressure drop (implying low energy losses).

A key aspect of the project, was the feedstock preparation for the selected auto-thermal reforming reactor. This activity showed difficulties connected with auto-ignition, coke formation and different corrosive properties of biodiesel in comparison with petroleum diesel. This article is focused at presenting the main result obtained in this context.

### 3. Feedstock conditioning

#### 3.1. Evaluation of the required biodiesel flow rate

The theoretical hydrogen flow to be fed to a 10 kW<sub>e</sub> PEMFC stack can be calculated using Faraday's law:

$$\dot{n}_{\text{H}_2} = \frac{i_{\text{stack}}}{zF} = \frac{P_{\text{stack}}}{zFE} \quad (9)$$

where  $i_{\text{stack}}$  and  $P_{\text{stack}}$  are, respectively, the current and the electric power of the FC stack,  $z$  the number of electrons transferred in the cell reaction,  $F$  the Faraday number, and  $E$  is the single cell voltage. The cell voltage was fixed to 0.6 V,

since this cell voltage gives a good compromise between cell efficiency and the stack dimension.

The ideal hydrogen flow rate according to Equation (9) is 0.0864 mol H<sub>2</sub> s<sup>-1</sup> (assuming a fuel utilization of 100%). The fuel utilization factor ( $U_f$ ) refers to the fraction of the total fuel (H<sub>2</sub>) introduced into a fuel cell that reacts electrochemically. In the PEMFC, the evaluation of  $U_f$  is relatively straightforward, because hydrogen is the only reactant involved in the electrochemical reaction. Usually,  $U_f$  is about 80% in the PEMFC, thus, in the outlet stream 20% of the fed hydrogen has to be accounted for. Secondary reactions, such as current leakage and crossover streams cause further limited hydrogen misuse, so that the fuel utilization  $U_f$  can be set at about 78%. Hence, the hydrogen flow required by the FC stack to generate 10 kW<sub>e</sub> is 0.11 mol H<sub>2</sub> s<sup>-1</sup>.

The biodiesel average molecular formula is C<sub>18.96</sub>H<sub>35.64</sub>O<sub>2</sub> and its average molecular weight is 295.7 [9]. Since the hydrogen flow rate is partially burnt in the CO-PROX unit owing to a selectivity hardly exceeding 50%, an excess of fuel is required to guarantee the hydrogen flow rate required by the FC stack. However, the total amount of biodiesel also depends on the production process conditions. For an ATR reactor, under stoichiometric conditions, the ideal auto-thermal conditions are: 1 biodiesel/21.6 air/26.8 water (molar ratio). However, a large excess of water must be used to decrease the CO content and for coking prevention. Excess water can be fed with a water injection (WI) system partly into the ATR reactor, to prevent coke formation, and partly after the reactor to displace the WGS equilibrium. An excess of air must also be used to decrease methane (and other unburned compounds) formation and to increase the reactor temperature so as to avoid risks of reaction extinction.

By means of an ad hoc fuel processor simulation tool (Matlab/Simulink platform), the heat and mass flows in the entire system were evaluated. A large number of runs have been carried out for a variety of inlet conditions to evaluate the optimal operating conditions at the system level (CO and CH<sub>4</sub> contents in the reformer outlet gas, H<sub>2</sub> yield, temperatures and biodiesel flow). The steam/carbon ratio in the inlet stream was finally set equal to 2, and the air/biodiesel ratio was tuned to have a reformer outlet temperature of 710–720 °C. The feed conditions fulfilling these requirements are: 1 biodiesel/35 air/38 water (molar ratio); the biodiesel flow rate required to guarantee the hydrogen flow in the fuel cell stack was, thus, evaluated to be 3.0 mmol H<sub>2</sub> s<sup>-1</sup>.

#### 3.2. Selection of feed strategy for the ATR system

According to the model calculations and the experimental data supplied by the catalyst developer (Johnson–Matthey) for the selected ATR catalyst composition, the gaseous mixture of water, air and biodiesel has to be fed into the reformer at 450 °C and 2.5 bar of absolute pressure. This condition can be reached by several means. However, some constraints must be considered concerning coke formation and

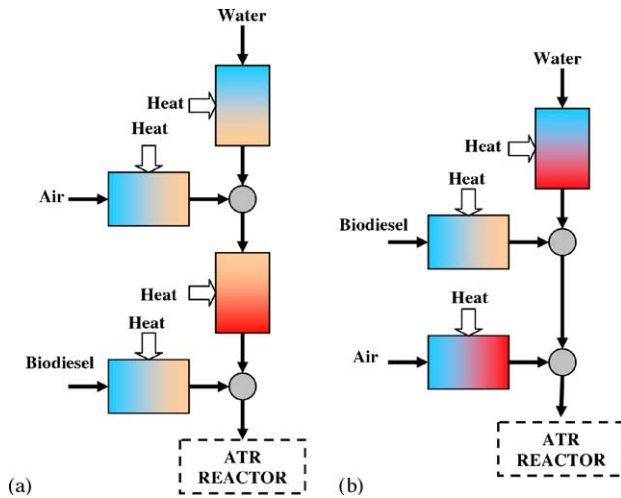


Fig. 2. Possible feed sequences for the ATR reactor.

avoidance of auto-ignition of the feed mixture before reaching the catalyst.

The first studied feed sequence (Fig. 2a) involved the vaporization/overheating of water and the air pre-heating at least at the same temperature of steam before the air injection to avoid the water condensation. According to this scheme, the flows of water and air are larger than the flow of biodiesel, therefore, the heat held in the mixed stream is enough to evaporate all the biodiesel, which could be sprayed by a nozzle or evaporated using a coiled tube evaporator.

According to a second scheme (Fig. 2b), steam is produced and overheated and the fuel is injected as liquid phase through a nozzle or evaporated by the steam flow using a coiled tube evaporator. The steam flow temperature must be enough to evaporate all the biodiesel. Air is pre-heated and added to the steam/fuel mixture after that.

This last approach was chosen for the final BIOFEAT prototype owing to two major advantages over the alternative route:

- Biodiesel injection in a pure steam flow was found to reduce the effects of cracking and coke formation via promotion of coke-steam reactions leading to CO and H<sub>2</sub>.
- Air is injected just at the very last moment ahead of the ATR reactor, with reduced risk of explosive mixtures formation and/or ignition.

### 3.3. The selected biodiesel feeding solution

Different fuel feeding strategies can be applied to the ATR reactor. The first considered option is to use an *oil nozzle*, like those used in high-pressure oil burners; the fuel is mixed with steam using a cyclone effect in a properly designed mixing chamber. As earlier mentioned, air is added immediately before the ATR reactor, and the mixing of the steam/fuel mixture with air is ensured by the expansion of the gas stream entering the reformer.

The second option is to use the steam flow to vaporize the fuel pumped in a coiled stainless steel pipe evaporator. The biodiesel vapours are then mixed with steam at the tube outlet. As well as for the other solutions, air is added to the steam/fuel mixture just before the reformer inlet. This solution assures a good dispersion of biodiesel into the steam flow. Moreover, the diameter of the pipe (1/8 in.), comparatively large if compared with the nozzles size, plays in favour of a reduced possibility of plugging due to coke formation.

The selection of the best fuel feed solution was based on a number of criteria; the most important are *stability, compactness, probability of plugging, sensitivity to vibrations, emissions, start-up time, turn down ratio*.

The flow rate stability turned out to be the most important requirement for the fuel supply system; a fluctuating or not stable fuel flow involve fluctuations in the reformat composition and in the reactors temperature, making the overall system less controllable. From this point of view, the use of a coiled evaporator is risky, because of intrinsically fluctuating behaviour of the evaporation process.

Moreover, the evaporator requires a greater mixing chamber than the nozzle, to accommodate the coil itself. So, the nozzle system would be more compact.

Another fundamental requirement for the feed unit is the low probability of plugging due to coke formation. The greater diameter of the pipe (compared with the nozzle), probably, provides a reduced possibility of plugging. Nevertheless, experimental characterizations by the Politecnico di Torino showed plugging of the pipe after some hours of operation with biodiesel.

Furthermore, since the fuel processor is designed for an automotive application, a good shock and vibration resistance is required. This feature is assured by the nozzle solution; on the contrary, the coil evaporator could be damaged in a real automotive application if not properly welded to fixed structures along its entire length.

Moreover, the BIOFEAT fuel processor is characterized by reduced noxious emissions, but in the case of the coil evaporator, during shutdown the evaporation of the fuel would continue for some time, causing the emission of uncombusted hydrocarbons.

As far as the system start-up time is considered, transients are shorter for the nozzle injector, because evaporation of the fuel in the coil is slowed down by the heat capacity of the pipe.

Some final consideration must be given to the turn down ratio of the fuel processor, which has been fixed equal to three. The nozzle solution requires changing the pressure applied to the injector over a wide range, to obtain a wide flow rate range (see later in Section 4.2). On the contrary, for the evaporator, the required fuel flow can be obtained with a simple flow controller placed upstream.

As with the previous considerations, the nozzle solution was selected for the BIOFEAT fuel processor. The problems related to the limited turn down ratio, a weak point of



this solution, were solved by a dual nozzle feed system as explained in Section 6.

## 4. Experimental

### 4.1. Experimental equipment

The equipment for the first lab tests were selected with low-cost high-pressure oil burner components (based on copper alloys). A simple lab-scale system was built to test single components, such as oil nozzles, valves and pumps with biodiesel. During the first tests, the fuel was pre-heated to decrease viscosity and improve spray dispersion. In a second phase, the fuel was not pre-heated but the nozzle was kept to high temperature (up to 250 °C), and the cold biodiesel flow was used to cool down the nozzle and avoid coke formation and plugging. The used oil nozzles, supplied by the company “Hago”, are entirely made (except for the porous filter integrated, Fig. 3) using a special heat and corrosion resistant high chrome stainless steel. The tested nozzles were “ES – solid cone – green caps” with various nominal flow rates (from 0.3 to 1 gal h<sup>-1</sup>, 1 GPH = 1.1 × 10<sup>-6</sup> m<sup>3</sup> s<sup>-1</sup>) and cone angles 30°, 45° and 60°.

Scanning electronic microscopy (SEM) analyses were performed on filtering materials using a Philips M525 30 KV microscope. Inductive coupled plasma technique (ICP), performed by a Perkin-Elmer OPTIMA 3000 instrument, was used to analyse the biodiesel fuel before and after operation in the fuel line.

### 4.2. Pressure flow rate relation

The first characterization performed on oil nozzles was devoted to obtain the pressure/flow rate curve. According to [10], the relation between mass flow rate and applied pressure is:

$$F = F_R \sqrt{\frac{P}{P_R}} \quad (10)$$



Fig. 3. Hago oil nozzle: the component is made using high resistance stainless steel except for the sintered bronze filter.

where  $F_R$  is the reference mass flow rate for the nozzle at the reference pressure  $P_R = 100 \text{ PSI} = 6.89 \text{ bar}$ . The injection chamber in all tests was not pressurized, so the pressure  $P$  in Equation (10) is the relative to atmospheric pressure.

## 5. Results

### 5.1. Nozzle testing

The experimental curves between the mass flow rate and pressure for the 0.85 and 1 GPH nozzles are reported in Fig. 4. The most relevant finding is that, with no fuel pre-heating and with an applied pressure lower than 4 bar, there is no spray formation but only dripping of large drops from the nozzle. With fuel pre-heating at 70 °C, spray formation can be obtained even at a lower pressure (2 bar).

As mentioned earlier, other types of tests involved the direct heating of the nozzle with no fuel pre-heating. In this case, the injection chamber was filled with nitrogen to avoid auto-ignition. This simulates the actual working condition in the fuel processor prototype. The nozzle was heated up to 250 °C with no evidence of plugging. Probably, the short contact time between the fuel and the hot metal of the nozzle, did not allow fuel degradation.

### 5.2. Corrosion problems

The low-cost oil burner filter components chosen for the lab tests are built using copper and copper alloys. The use of copper alloys caused corrosion problems and, as a consequence, a possible pollution of the fuel with copper ions.

Inductive coupled plasma analysis (ICP) showed that the copper content in the used biodiesel is 21 ppm after 2 h of operation, against 0.1 ppm for the original fuel.

Copper traces in the fuel can negatively affect the catalysts used in the chemical reactors of the fuel processor and have to be avoided.

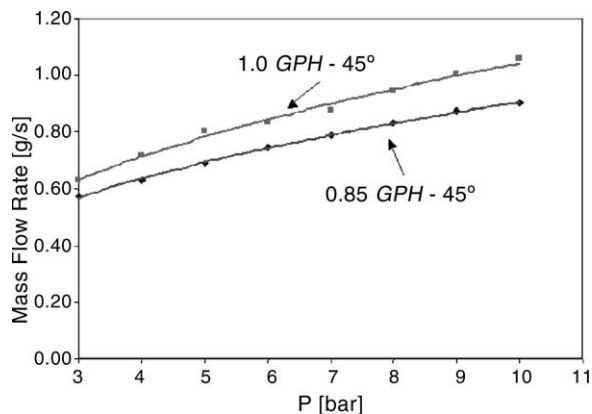


Fig. 4. Mass flow rate vs. pressure relation for the 0.85 and 1 GPH oil nozzle at 70 °C. Spray formation at low pressure can be obtained only with fuel pre-heating.

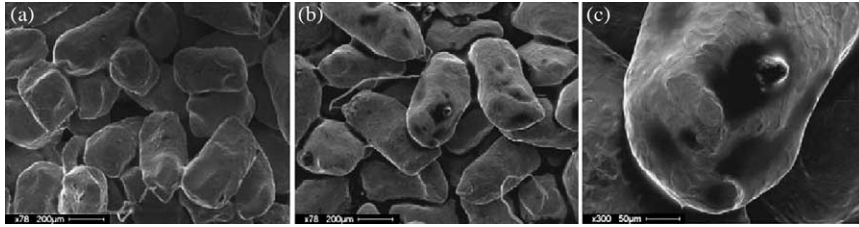


Fig. 5. SEM image of the new (a) and used (b and c) bronze filter. Pitting corrosion appears after several hours of operation with biodiesel at 70 °C.

Corrosion signs are evident on the bronze sintered filters integrated in the oil nozzle after 10 h of operation with biodiesel at 70 °C (Fig. 5).

## 6. Discussion

The experimental tests and analyses performed provide some useful considerations in the construction of the final BIOFEAT feed system prototype.

The corrosion tests, showed the need for copper-free components; in particular, the oil pump and the oil filter have to be stainless steel and cannot burrow from the oil burner technology (in which copper alloys are widely used, because copper pollution does not affect fuel combustion). For the final fuel processor, a new stainless steel metering pump and sintered stainless steel filters were selected.

Despite the good turn down ratio, the drawbacks of the coiled pipe evaporator were considered too serious for safe application in a biodiesel fuel processor. On the other hand, the oil nozzle configuration requires further development to reach a good turn down ratio. As a matter of fact, to regulate output power from 3 to 10 kW, supposing an operating pressure of 2.5 absolute bar for the ATR reactor and using a nozzle with a nominal capacity of 0.55 GPH, the biodiesel feed pressure should be varied (according to Equation (10)) from 4.5 to 30.5 absolute bars. On the other hand, the required flow rate is very low and commercial stainless pumps that can reach the needed maximum pressure give a too large mass flow rate at these pressures, so the major part of the pumped fuel has to be re-circulated using a by-pass circuit. As a consequence, the use of an over-dimensioned pump would not be convenient from an energetic point of view (due to the greater electric power required).

Possible solutions for the regulation of the fuel flow are the use of a *pulsed metering valve* or the application of two nozzles each with a nominal capacity half the required maximum fuel flow rate. The metering valve is a high-cost solution, applied in previous liquid fuel processor projects [5], but available commercial valves require further material development to withstand biodiesel corrosive properties. The other simple and low-cost solution is the use of two independent nozzles controlled by electro-valves. The first nozzle is opened from start-up up to 5 kW, the second is used when the required power exceeds half the nominal power of the APU. Moreover, the use of electro-valves avoids

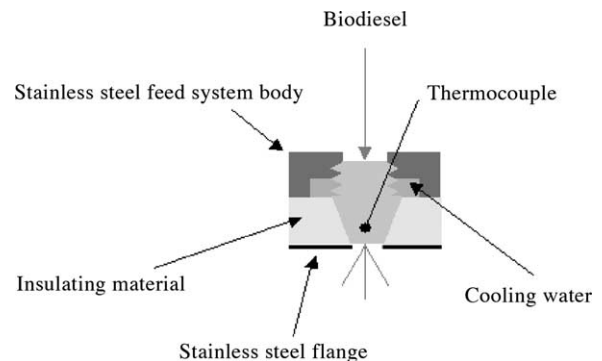


Fig. 6. Biodiesel nozzle cooling system. The nozzle tip temperature has to be controlled by the system control system and kept below 250 °C.

possible dripping after pump shutdown, which could result in occurrence of noxious emissions.

The last important point for the design of the biodiesel feed system lies in the need for a proper cooling of the nozzles, through insulating materials placed on the nozzle itself and a water-cooling system. The nozzles are in fact in contact with the steam flow that is fed in the mixing chamber at 600 °C. The fuel feed is not pre-heated and is used to further cool the nozzle tip. To avoid plugging and coke formation, the temperature of each nozzle has to be measured and kept (acting on the cooling water flow rate) below 250 °C. A detailed view of the nozzle cooling system is shown in Fig. 6.

## 7. Conclusions

The feedstock conditioning issue for an automotive fuel cell auxiliary power unit based on a biodiesel processor has been studied. The components and materials testing (corrosion) allowed selection of the proper commercial components suitable for the project operating condition and the chosen fuel. The nozzle testing activity supplied useful information for the project of the final feed unit prototype, which will be based on a dual nozzle system controlled by suitable electro-valves and including entirely copper-free components.

## Acknowledgments

The authors gratefully thank the European Commission for funding this project within the V Frame Programme (contract number: ENK6-CT2002-00612). Special thanks are due

to all the other partners involved in this research project: Energy Research Centre of the Netherlands (NL), University of Duisburg-Essen (D), Bekaert VDS (B), Johnson Matthey (UK), Scandiuzzi Advanced Technologies (I).

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