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Biogas as a fuel source for SOFC co-generators

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Abstract

This study reports on the combination of solid oxide fuel cell (SOFC) generators fueled with biogas as renewable energy source, recoverable from wastes but at present underexploited. From a mobilisable near-future potential in the European Union (EU-15) of 17 million tonnes oil equivalent (Mtoe), under 15% appears to be converted today into useful heat and power (2 Mtoe).

SOFCs could improve and promote the exploitation of biogas on manifold generation sites as small combined heat and power $(5-50 \text{ kW}_{el})$, especially for farm and sewage installations, raising the electrical conversion efficiency on such reduced and variable power level. Larger module packs of the high temperature ceramic converter would also be capable of operating on contaminated fuel of low heating value (less than 40% that of natural gas) which can emanate from landfill sites (MW-size). Landfill gas delivers 80% of current world biogas production.

This document compiles and estimates biogas data on actual production and future potential and presents the thermodynamics of the biogas reforming and electrochemical conversion processes. A case study is reported of the energy balance of a small SOFC co-generator operated with agricultural biogas, the largest potential source.

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1. Introduction

Biomass offers world-wide the largest exploitation potential among renewable energy sources. In the European Union, bioelectricity production is especially encouraged, a significant portion of which could be met alone from biogas.

Biogas fuel feeding presents an attractive option among emerging applications for fuel cells, especially for the high temperature ceramic type solid oxide fuel cells (SOFCs). Compared to natural gas (NG), it shows advantages of being indigenous and renewable, free of non-methane hydrocarbons (NMHC), with the exception of landfill gas and containing a large fraction of a methane-reforming agent (CO₂). Biogas fabrication inherently is a friendly and senseful way to process waste streams of variable nature (sewage sludge, liquid organic industrial effluents, farm residues, landfill, municipal and industrial solid organic residues).

A general problem with biogas resources is their local nature. Residues from farms and municipalities typically represent small power houses in the range of $5-100 \text{ kW}_{el}$. Especially the smallest sources, when used with engines or microturbines, show low electrical conversion (20%) and high levels of noise and maintenance requirement. Exploitation for such sources rather involves large farms or waste collecting schemes (>10,000 t per year) to large digester installations, coupled to conventional engines of the 1 MW_{el} size, where these present a reasonable efficiency (35%).

A particular problem with some of existing biogas installations (landfill gas (LFG) now representing 80% of the world biogas production) is that dilute gas remains unused. Landfill sites may emanate poor quality gas, containing toxics like halogenated and sulphurous compounds and being of low calorific value (<40% CH₄). A 1997 EU regulation specifies to reduce organic matter in European landfill by 75% until 2010 [1], which is bound to dilute the methane content in LFG. Conventional technology stops operating on such fuel. In some cases even fossil fuel-assisted flaring of the off-gas is employed, in order to avoid methane venting.

SOFCs could be an appropriate conversion technology for biogas, achieving reasonable efficiencies (30–40%) already in the smallest power range (5–20 kW_{el}), being safe, silent and expected to be low in maintenance. It may probably be

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ity biogas from landfill. In laboratory tests, high electrical conversion was maintained down to very low methane levels (5%) [2], and performance drops of only 5% were registered when operating on mixtures of 30–70% CH₄–CO₂ compared to 70–30% CH₄–CO₂ [3]. The first part of this paper compiles biogas production data in Switzerland and Europe to estimate the exploitation potential by SOFCs. The second part considers the energy balance model on an existing small SOFC co-generator fed on agro-biogas, where the largest exploitation potential lies.

2. Discussion

2.1. Biogas sources

We are concerned with biogas generation from methanisation, by digestion or landfilling. Application examples are given in Table 1. The proportion of dry organic matter in these wastes is often too low for incineration to be efficient, so that anaerobic digestion presents the method of choice. Considered sources indicated below are ranged from more liquid to more solid nature:

- Industrial waste waters (WW) with a high organic charge (<5% dry matter) from the agro- and food industry: milk, cheese, fruits, vegetables, meat, fats, sugars, bakeries, potatoes, pastas, rice, coffee and tea, breweries and others like slaughterhouses, paper/pulp, cosmetics, pharma, oil. The alternative to digestion is oxygenation and disposal. In waste water treatment plants, half of the co-generated heat is used to sustain the methanisation process.
- Municipal waste water residue or sewage sludge (<5% dry matter). The alternative to digestion is oxygenation or incineration. The former involves a high chemical and biological oxygen demand (COD, BOD), which is energy intensive because of pulsed oxygen injection (1 GWh per year for an average waste water treatment plant). The resulting sludge after aerobic purification has not experienced an important volume reduction (20,000 t sludge per year for an average waste water treatment plant) and still requires post-elimination (incineration, landfill) [4] Anaerobic degradation produces more output (biogas) than the energy it requires to run the process and reduces

sludge output to 5% of the initial volume. The output is fertilizer, completing a sustainable cycle.

- Livestock (animal manure) and green waste (straw and harvest residues) on farms (<10% dry matter). When not digested, this is recycled by aerobic composting. Sixty cattle (i.e. 20 kW_{tot}) is a minimal critical size [5].
- Organic fraction of solid wastes, i.e. fruits, vegetables, plants, paper, grass (~30% dry matter of which ≈75% organic, for a useful fraction of 22%): households, restaurants, supermarkets, slaughterhouses, paper industry, chemical industry, public zones (parcs, etc.). Such waste streams are treated in centralised, dedicated sites (digestion, landfill, incineration). Fermentation being economic above a critical mass of 50,000 inhabitants, the minimal size for such digestion plants is around 500 kW_{tot} [5], whereas incineration is based on an economy of scale (MW sized plants). Where incineration costs € 200/t for a volume reduction factor of 4, landfilling costs € 60/t for a biogas production of 100–200 m³/t waste during its 10–20 years of operation.

Note from these data (Table 1) the variable annual load factor, the efficiency (18–36% electrical), and the site power size. Power rating of $10-100 \, kW_{el}$ (farms, sewage) to $0.1-1 \, MW_{el}$ per site (solid waste digestion and landfill) is typical. This corresponds to market niches for fuel cell systems, where turbines show lower efficiency and engines higher (chemical and acoustic) pollution values. Fuel cell units are scaleable and modular, adaptable to the site requirements between 5 kW and 1 MW, advantageous for CHP penetration.

2.2. Data on biomass, bioelectricity and emissions

2.2.1. Biomass and biogas

The biomass share in 1995 in the European Union (EU-15) was 3% or 45 million tonnes oil equivalent (Mtoe) (1 Mtoe = 44 PJ). This primary energy was composed of 85% woods and 15% non-woods (energy crops, incinerated solid wastes, biogas), to give 5% of the final energy and just 1% of electricity production (22.5 TWh). On a world-wide scale, the biomass share lay at 8% (1995). By 2010, the EU-15 wants to convert 12% of its primary energy from renewable sources, the major part originating from biomass (135 Mtoe or 8.5%). The additional 90 Mtoe of biomass are

Table 1

Existing application examples of biogas production plants (Switzerland)

Example	Produced biogas (m ³ per day)	CH ₄ (%)	Site size (kW _{tot})	Electrical power (kW)	Annual load (%)	Electrical efficiency (%)	Heat used (%)	Reference
Farm 37 cattle	70	57	17	5	60	18	49	[6]
Farm 104 cattle	260	62	70	37	55	29	n.a.	[7]
Sewage plant 30000 habitants	1000	65	265	130	65	28	n.a.	[8]
MSW 80000 habitants	1300	60	340	90	95	25	60	[9]
MSW 350000 habitants	3800	55	918	350	95	36	27	[10]

MSW: municipal solid waste digester plants.



Fig. 1. Schematic of existing (1995) and planned (2010) biomass use in Europe.

planned to be mobilised as [1]—see Fig. 1: 30 Mtoe from more wood, 45 Mtoe from new energy crops and 15 Mtoe from better biogas exploitation, namely from sewage, livestock, landfill and solid residues (agro, industrial and municipal). Over 50% of this new biomass can be used in combined heat and power production, CHP (32 Mtoe from wood and crops, 15 Mtoe from biogas). The ultimate total biogas potential (EU-15) is estimated to 80 Mtoe per year [1].

2.2.2. CO_2 emissions

The EU-15 CO₂ emission in 1990 was 3 Gt (world: 21.6 Gt) and originates from [1] power generation (33%), transportation (20%), industry (18%), the domestic sector (17%) and the energy sector (10%). According to the Kyoto Protocol target, this number must be reduced by 8% to 2.75 Gt per year in 2010. The expected emission in 2010, following present trends, would amount to 3.55 Gt per year. The required reduction is thus 800 miot CO₂ per year.

Fossil fuel conversion (EU-15) in 1999 was 1.138 Gtoe (coal 209 Mtoe, oil 587 Mtoe, gas 340 Mtoe). With carbon emission factors of 1.47, 0.844 and 0.602 Mt C/Mtoe for coal, oil and NG respectively, the mixed emission factor for Europe is derived as 0.885 Mt C/Mtoe. Replacing fossils by 90 Mtoe CO₂-neutral biomass fuel then theoretically avoids 80 Mt carbon emission, or 290 Mt CO₂.

Thermally generated electricity (EU-15, 1999) was derived from 276.5 Mtoe fossil fuels (151 Mtoe coal, 35 Mtoe oil, 80 Mtoe gas). With 1347 TWh produced, the average generation efficiency was 40%. The accompanying mixed emission factor is calculated to 1.13 Mt C/Mtoe or 223 t C/TWh_{el} (816 t CO₂/TWh, 1.1 Gt CO₂ per year).

Thus, when producing electricity from CO_2 -neutral biomass rather than from the existing fossil fuel mix, even without co-generation, an emission saving of 0.82 kg CO_2 per kWh is obtained.

2.2.3. CH₄ emissions

Exploitation of biogas, especially at landfill sites, improves control over methane leakage. Methane is a major greenhouse gas, 30 times more powerful (in molar terms) than carbon dioxide. The greenhouse effect on the planetary scale is indicated, depending on source [11–13], to arise from partial contributions of CO₂ (50–55%), CH₄ (7–19%), CFC (17–28%), N₂O (7%) and others like O₃ (3–7%).

Atmospheric methane, the fraction of which has risen from 1 in 1920 to 1.7 ppm in 1990 [11], originates from both anthropogenic and natural emission. There appears consensus on the following data:

- Thirty percent of methane emission is natural (from wetlands, termites, oceans, freshwater, gas hydrates) and 70% is anthropogenic (from fossil fuel mining, livestock, rice farming, biomass burning, landfilling, etc.);
- the yearly atmospheric methane excess is around 40 Mt C (roughly 90% of total methane emission is destroyed by atmospheric reactions with OH-radicals and a smaller fraction—5%—by soil oxidation);
- landfilling accounts for an average emission of 40 Mt C per year, livestock account for an average emission of 80 Mt C per year.

Therefore, by intense exploitation of biogas from livestock and landfill, it seems that the atmospheric methane increase could be entirely suppressed, and with it around 10% of the greenhouse effect.

2.2.4. CHP

In 1995, the EU produced 2366 TWh electricity from 549 GW_{el} power, with a load factor of 50%. In terms of CHP, 67 GW_{el} (13% of installed power) was in operation, for an electricity production of 205 TWh_{el} (9% of produced power), and a load factor of only 35%. The main

EU-15	1995 data			2010 forecast		
Electricity	Installed (GW)	Load (%)	Produced (TWh)	Installed (GW)	Produced (TWh)	Increase (1995–2010)
Total	549	50	2366	>600	2870	1.21*
CHP	67	35	205 (9%)	>134	516	2.5*
Biomass	5 ^a	50 ^a	23 (1%)	50 ^a	230	10*

Table 2 Electricity production data and forecast in the EU-15

^a Estimated.

* = multiplication sign.

CHP objective [14] is to double this share to 18% in 2010 (516 TWh_{el}), mobilising an additional 300 TWh_{el} of CHP electricity.

Reaching this goal would massively contribute to CO₂ emission reduction. The mixed emission factor of 0.885 Mt C/Mtoe corresponds to 265 kg CO₂ per MWh of CHP fuel, that is assumed to be converted to 0.4 MWhel (40% average conversion efficiency from fossil fuels) and 0.40 MW heat with a total efficiency of 80%. When separately generating the same amount of electricity and heat from fossil fuel (at 40 and 90% efficiency, respectively), the associated CO₂ emission will be 265 kg (for 0.4 MWh electricity) and 118 kg (for 0.40 MWh heat). The cumulated saving per MWh of CHP electricity is then 118/0.4 = 295 kg CO₂. The major part of the target (>300 TWh_{el} new CHP) can be achieved through biomass (additional 200 TWhel in 2010) [1], as summarised in Table 2. The maximum CHP potential (EU-15) is estimated to level out at 1000 TWh per year (over one-third of the total electricity).

2.3. Biogas data

Relevant data for Switzerland [15] are compiled in Table 3 and data pertinent to Europe (EU-15) are tentatively summarised in Table 4.

Note that the electrical efficiency figure is the product of the actual efficiency with the (unknown) annual load factor. The primary purpose of waste water (WW) treatment plants is detoxification, hence a large fraction of heat is recovered (60%) for plant operation and electrical efficiencies are not optimised. Contrarily, the main byproduct of solid waste treatment is electricity, a large fraction of heat is rejected unused (60%). The size of biogas power sites (see also Table 1)—assuming 30% efficiency and 100% load—lies at around 500 kW_{el} for landfill, 100 kW_{el} for waste water residues and solids digester plants and 10 kW_{el} for farms, compared to several MW_{el} for a waste incineration plant, in the case of Switzerland. Use of wood energy lies higher than biogas by an order of magnitude (similar to Europe), and elimination of the renewable part of solid wastes is mostly incinerated, else landfilled, with only the smaller fraction being digested. Total actual biogas production is around 115 Mm³ per year, poised to grow to 300 Mm³ per year until 2010 [5].

Apparent efficiencies are <20% for biogas conversion. For industrial waste waters (no direct data), the figure for Switzerland (0.132 PJ) is extrapolated to EU-15. The solids digestion data take into account the known 105 sites that each process more than 2500 t waste per year for a total of 4.18 Mt per year [18]. Biogas generation was taken as 130 m^3 /t waste [9]. The average energy capacity per site is 3.4 MW total, or roughly 1.25 MW_{el} installed, when assuming 35% efficiency and a 95% annual load. Site capacity for the indicated 700 agroinstallations [16] would amount to 5.5 TJ or 176 kW_{tot} (refering to large farms of >400 cattle, ca. 50 kW_{el}/site) but only 13.4 kW_{tot} per sewage digester, where several digesters are combined in a single plant.

In total, 150 Mt per year solid wastes (municipal, industrial) are generated in EU-15, of which 2/3 end in landfill (99 miot per year), 2/9 in incineration (33 miot per year) and the remaining 1/9 (16 miot per year) in recycling, composting or digestion (4.2 Mt per year) [17]. Summarised, just 2 Mtoe (87 PJ) of biogas primary energy is estimated to be exploited now, with ca. 60% in landfill, up to a quarter in waste waters and the remainder in solids digestion; agricultural biogas, potentially the largest source, contributes with less than 5%. The current world biogas production is

Table 3

Renewable er	nergy statistics	for Switzerland	1 (2000),	especially	biogas
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Source	Brutto (PJ)	Electricity	Used heat	Sites	kWtot per site
Municipal WW (sewage)	1.736	98 GWh (20.3%)	0.956 PJ (55%)	285	193
Industrial WW (organic effluents)	0.132	2 GWh (5.7%)	0.087 PJ (66%)	18	232
Solids in landfill	0.573	44 GWh (27.7%)	0.077 PJ (13.4%)	13	1397
Solids in digesters	0.111	6.7 GWh (21.8%)	0.0188 PJ (17%)	11	319
Livestock	0.061	3.25 GWh (19.3%)	0.0122 PJ (20%)	68	28
Total biogas	2.613	154 GWh (21.2%)	1.151 PJ (44%)	395	
Renewable fraction of Incinerated solids (municipal)	16.786	634 GWh (13.6%)	4.38 PJ (26.1%)	28	18997
Renewable fraction of Incinerated solids (industry)	2.762	36 GWh (4.7%)	2.06 PJ (74.5%)	37	2365
Woods	19.6	14 GWh	12.2 PJ (62%)	650k	

Table 4 Estimated renewable energy statistics for Europe, especially biogas

Source	Production (Mm ³ per day)	Sites	CH ₄ (%)	Total (PJ)	Electricity (GWh)	Reference
Sewage	1.7	36000 digesters	65	15.2	650 (15%)	[16,17]
Industrial WW	0.8^{a}	400 installed	65	7 ^a		Estimated [17]
Landfill	7.2		50	50	1351 (10%)	[17]
Solids digestion	1.5	>105	55	11.25 ^a	1000 ^a (36%)	[18]
Livestock	0.5	700	55	3.8	133 (12%)	[16,17]
Total biogas	11.7			87		
Solids incinerated				180 (36% heat)	15000 ^a	[17]
Woods				1700 (54% heat)	3525	[17]
Total biomass				45 Mtoe	22500	

^a Estimated.

believed to lie at 52 bio m^3 per year (140 Mm³ per day), or 1.3% of actual natural gas production, of which 80% is generated in landfill [16].

2.4. Biogas potential

Exploiting the ultimate biogas potential for Switzerland would lead to the following numbers [4,17].

For sewage, on average 301 per day of biogas (65% CH₄) per person-equivalent (p-eq.) is generated in working installations [8] (see Table 1).

Industrial wastewaters amount to ca. 21 mio m^3 per year effluents, with ca. 1% organic dry matter (ODM) [4]. With 0.45 m³ biogas (65% CH₄) generated per kg ODM, this potential translates to 13.5 m³ per year p-eq.

Solids wastes from municipalities (household, public zones) total 1.08 Mt ODM per year (17 PJ) and from industry 0.52 miot ODM per year (8.7 PJ) [17]. Most of this is currently incinerated (19.5 PJ, Table 3), the remainder being landfilled, composted or recycled. A significant fraction (180,000 t per year ODM from household, 50,000 t per year from industry, 60,000 t per year from public zones) could be digested, to ca. 3 PJ of biogas [4]. Hence, 125 kg per year p-eq. of digestable household waste (27% fraction of the total household waste) yields 12.375 m^3 per year p-eq. of biogas (60% CH₄, 22% ODM, 0.45 m³ biogas/kg waste, or 100 m³ biogas/t waste) and other solid wastes equal 17 kg ODM per year p-eq. or 7.65 m³ per year p-eq. biogas (60% CH₄, 0.45 m³ biogas/kg waste).

Livestock could ultimately derive 4.04 miot ODM per year or theoretically 63.6 PJ in one estimate [17]. Using a simple and more moderate approximation by considering cattle statistics for Switzerland and applying tabulated conversion factors, a rough equivalent of 2 million cattle units is found which, rated at a daily biogas production of 1.5 m³ per day, yield a total of 1.1 bio m³ per year or 22.7 PJ (55% CH₄). Solid residues from harvesting and straws, co-digested with the liquid manure, would yield another one-third of the livestock potential [17], or 7.6 PJ.

Summarising these sources (Table 5), a theoretical yearly biogas potential of 57 PJ for Switzerland results (6% of the country's primary energy).

Table 5						
Estimate	of	theoretical	biogas	potential	for	Switzerland

Source	Energy (PJ)	CH ₄ (%)	Equivalent (m ³ per year p-eq.)
Sewage	1.9	65	11
Industrial WW	2.3	65	13.5
Solids household	1.8	60	12.375
Solids other (industry, public)	1.1	60	7.65
Solids now incinerated	19.5		
Solids in LF	0.5	55	
Livestock	22.7	55	
Solid agroresidues	7.6	55	
Total	57.4		44.5

For a realistic potential, figures for sewage waste wasters (1.9 PJ, see also Table 3), solids (2.9 PJ) and agriculture (10% of total of 30.3 PJ, or 3 PJ) are assumed, to give ca. 8 PJ. Extrapolating this potential to the situation for EU-15 yields the estimate of Table 6. The livestock figure is derived from estimated 130 mio large cattle equivalents [17] (200 Mm³ per day, 71 bio m³ per year).

The realistic exploitable figure is that from sewage sludge (101 PJ), solids (206 PJ, of which now 50 PJ in landfill) and agriculture (10% or 196 PJ) for a total of ca. 500 PJ (11 Mtoe). The White Paper on Renewables [1] mentions a mobilisable potential of 17 Mtoe biogas by 2010, and an ultimate potential of 80 Mtoe, consistent in the order of magnitude with the numbers estimated here.

Table 6 Estimate of theoretical biogas potential for Europe

Source	Energy (PJ)
Sewage	101
Industrial WW	124
Solids household	96
Solids industry, public	60
Solids now landfilled	50
Livestock	1475
Solid agroresidues	491
Total	2400 PJ (54.5 Mtoe)

Biogas (17 Mtoe) reached only at a yearly biogas production increase of 20% until 2010, is equivalent to 11% of current EU-15 natural gas import. Exploited with a load factor of 80% in SOFC-CHP with 40% electrical and 45% thermal conversion, would imply the operation of 12 GW_{el} with a yearly production of 83 TWh_{el}, or 3% of EU-15 electricity, and 7.6 Mtoe of heat. Replacing this energy into the present European energy conversion mix (0.223 Mt C/TWh electricity, 0.885 Mt C/Mtoe heat) could avoid emissions of >90 Mt CO₂ per year (68 Mt per year from electricity, 25 Mt per year from heat).

3. Results

3.1. SOFC-biogas system case study

The choice of SOFCs compared to other fuel cell technologies presents distinct advantages. The high operating temperature (700–1000 $^{\circ}$ C):

- allows for co-generation at high temperature level and the elimination of noble metal electrodes and special fuel processing catalysts (e.g. shift) that are sensitivite to fuel impurities;
- allows thermal integration with the fuel cell stack of all fuel conversion and purification steps, thereby increasing the system efficiency.

SOFC modules in the range of 1 kW_{el} [19] to 1 MW_{el} [20] are being demonstrated or in construction and will therefore be able to cover all biogas power sites. Molten carbonate fuel cell (MCFC) modules are commercialised in 1 MW_{el} size and phosphoric acid fuel cells (PAFC) in 50–200 kW_{el} size [20], meaning they cannot cover many small agrosites, where the largest biogas potential lies (Tables 5 and 6). These technologies, of lower system efficiency than SOFCs, are also more sensitive to biogas impurities [21]. A system study comparing the different technologies is currently ongoing and will be published separately [22].

As a case study, the implantation of a small SOFC module on an agrosite is considered. The farm "Maison Blanche" in Lully, Switzerland, has exploited since 9 years [6] biogas production from livestock (37 cattle units), sometimes mixed with municipal waste from 1500 surrounding inhabitants, at a typical rate of 70 m³ per day (0.55 TJ per year or 17 kW_{tot}). It is equipped with a small co-generation gas engine of the total energy module (TOTEM) type, with 5 kW_{el} nominal and 21 kW_{thermal}. Electrical efficiency of this unit is 18% and thermal efficiency 73%; one-third of the heat is rejected. The annual load factor is 60% (Table 1).

Recently, a Sulzer HEXIS SOFC module (1 kW class) [19] has been taken in operation on the site, for demonstration and investigation purposes [3,23]. Due to cattle reduction and suspension of the municipal waste collection, biogas production has now been reduced to around half (9 kW_{tot}), still sufficient to satisfy the electrical and heating needs for the estate. In the following, a single SOFC unit large enough to exploit this site production (9 kW_{tot}) is modelled on its energy balance.

3.2. Reforming aspects

Two distinctions of biogas, compared to natural gas, are the absence of non-methane hydrocarbons (except for landfill gas) and the presence of a large proportion of carbon dioxide, both of advantage for fuel processing in SOFC.

Higher hydrocarbons are more prone to carbon formation than methane. At ambient pressure and standard concentrations, butane, propane and ethane decompose to the elements (hydrogen and graphite) already at 77, 113 and $197 \,^{\circ}$ C, respectively, compared to the considerably more stable methane, which thermodynamically decomposes to the elements only above 535 °C.

 CO_2 is a reforming agent, that will allow to partly convert methane into syngas for SOFC anodes. To avoid carbon formation with only CO_2 as reforming agent, however, an excess would be required far from the proportions as they are found in biogas. To operate in a thermodynamically safe region to avoid carbon deposition above $600 \,^{\circ}$ C, a 10:1 CO_2 :CH₄ ratio would be needed [23].

In order to avoid the formation of carbon with biogas as a fuel, other reforming agents have to be added. These can be water vapour or oxygen (air). For system simplicity, air is chosen in the present study.

The minimum quantity of oxygen (air) required to avoid carbon formation above 800 °C for a methane-rich biogas composition (2:1 CH₄/CO₂, see Table 1) was determined by thermodynamic calculus and the result plotted in Fig. 2.

For the specified conditions (800 °C, 2/1 CH₄:CO₂) an oxygen addition of 0.21 mol is required. Biogas can thus be mixed with air in a 1:1 volume ratio. For lower methane content in the biogas (see Table 1), the situation becomes even safer. The actual composition onsite the Lully farm estate has been monitored by gas chromatography [23] and found to approach 60:40 CH₄:CO₂. H₂S was found to vary



Fig. 2. Minimal O_2 addition (in mole) required to the biogas feed 0.666 CH₄ + 0.333 CO₂, so as to avoid carbon formation.



Fig. 3. Thermodynamic equilibrium concentrations (moles) in the temperature range 300-1000 °C for an initial feed of $0.6 \text{ CH}_4+0.4 \text{ CO}_2+0.21 \text{ O}_2$. The right hand side axis plots the corresponding open circuit voltage (OCV) or Nernst cell potential -E vs. air.

between 70 and 700 ppm and is removed by active carbon filters prior to admission to the reformer [3]. Fig. 3 and Table 7 show the thermodynamic equilibrium concentrations for a 60% CH₄ biogas: air co-feed in a 1:1 ratio (by volume).

At 800 °C, methane conversion is 98.2%, with syngas selectivity 89% and carbon selectivity 0%. Oxygen addition thus assists the biogas CO₂ fraction in reforming of methane to syngas without risk of carbon formation above 750 °C. Vice versa, the CO₂ fraction in biogas assists the partial oxidation with oxygen in methane conversion to syngas without danger of carbon formation. Combining methane with only oxygen (in absence of CO₂) in the same proportions (0.6CH₄ + 0.21O₂), also results in a large carbon presence over the whole temperature range considered. For the latter case, theoretical methane conversion is 94.1% (800 °C), with only 75.8% syngas selectivity and 31.8% carbon selectivity. This stoichiometry can be understood as $3CH_4 + O_2 \rightarrow$ $6H_2 + 2CO + C$.

The fuel feed of 0.6CH₄+0.4CO₂+0.21O₂ is compacted to the single chemical formula C₁H_{2.4}O_{1.22} (1 mol of fuel). The stoichiometric oxygen required for full combustion of this fuel is $n_{O_2} = C + (1/4)H - (1/2)O = 1 + 0.6 - 0.61 =$ 0.99. One mole of biogas-air co-feed fuel is then fully converted to carbon dioxide and water vapour by 0.99 mol of

Table 7 Numerical values from Fig. 3

<i>T</i> (°C)	400	500	600	700	800	900
CH ₄	0.2228	0.1765	0.1098	0.0547	0.0110	0.0012
H_2O	0.6225	0.5157	0.3645	0.1885	0.1261	0.1370
CO_2	0.2960	0.3300	0.3188	0.1974	0.1049	0.0843
CO	0.0055	0.0443	0.2179	0.6367	0.8841	0.9145
H_2	0.1320	0.3313	0.616	0.9020	1.0518	1.0606
C	0.4756	0.4492	0.3535	0.1112	0	0
-E/air (V)	1.0226	1.0223	1.0258	1.0403	1.041	1.0143



Fig. 4. Thermodynamic equilibrium concentrations at 800 °C as a function of fuel utilisation for the initial fuel feed $0.6 \text{ CH}_4 + 0.4 \text{ CO}_2 + 0.21 \text{ O}_2$.

oxygen. This allows to specify the quantities of oxygen required to convert a fraction of the fuel (fuel utilisation u_f) and so, using thermodynamic equilibria calculation, to determine the outlet gas composition as a function of u_f , as well as the fuel cell equilibrium potential (*E*, in Volt) at the outlet composition. The result is graphically represented in Fig. 4.

Part of the inlet oxygen is transferred to the anode side, leaving an empoverished oxygen stream at the cathode outlet. This oxygen utilisation will further diminish the exploitable cell voltage, especially when using a stoichiometric oxygen flow (with respect to the fuel flow) at the cathode. The effect is reduced by using an excess oxygen (air) stream at the cathode, expressed as λ (an integer multiple of the stoichiometric oxygen amount). For any λ , the cell voltage can be corrected as follows:

$$E = \frac{RT}{4F} \ln \left[\frac{P_{O_2}^{\text{cath}} (1 - u_{\text{f}} / \lambda)}{P_{O_2}^{\text{anode}}} \right]$$

The calculated result for various λ is shown in Fig. 5.



Fig. 5. Cell voltage (open circuit) as a function of fuel utilisation and excess air flow at the cathode (λ) for the fuel feed 0.6 CH₄ + 0.4 CO₂ + 0.21 O₂ (800 °C).



Fig. 6. Schematic overview of flows and heat exchanges (HEX) in the SOFC-biogas system.

Operating the fuel cell with stoichiometric oxygen ($\lambda = 1$) would, at 800 °C and 80% fuel conversion, result in an additional cell voltage loss of 37 mV, or 4%. Already by doubling the oxygen (air) flow ($\lambda = 2$), this loss is reduced to only 12 mV. For reasons of stack cooling it is customary to work at a three-fold air excess ($\lambda = 3$). The loss due to oxygen conversion is then, for this example, reduced to 7 mV or less than 1% loss of the theoretical cell voltage.

The stoichiometry of the complete chemical reaction taking place in the SOFC stack fuelled with the 60% CH₄ biogas-air co-feed (1:1) with $\lambda = 3$ equals

 $0.6CH_4 + 0.4CO_2 + 3.1814O_2 + 12.02N_2$

 $\rightarrow 1.2 H_2 O + 1 C O_2 + 1.9814 O_2 + 12.02 N_2.$

In order to try and exploit the current biogas production on the agrosite (around 0.01 mol/s CH₄, or 8.9 kW HHV) with a single SOFC module of Sulzer HEXIS design, the following assumptions were made:

- 1. Anode supported electrolyte cells are used, aiming at stack power densities of 0.35 W/cm² at 800 °C [24].
- 2. A sufficient number of cells is used (100 cells of 100 cm² each).
- 3. Fuel reforming is achieved with CO₂ from biogas and added air. No steam is used. The fuel feed is pre-heated to operating temperature with stack heat.

- 4. The (endothermal) reformer is kept at stack operating temperature using stack heat. This can be achieved by an arrangement of stack-loaded ceramic tubes filled with reformer catalyst [25].
- 5. Air is pre-heated in three stages (Fig. 6): (i) to a temperature of <500 °C by the hottest exhaust gas; (ii) to ca. 700 °C by an afterburner zone; and (iii) to operating temperature by excess stack heat. Anticipating on some of the calculations, the exhaust reaches 900 °C in the post-combustion zone, is cooled to around 550 °C for air pre-heating, finally used for domestic hot water production.

The flow system is schematically shown in Fig. 6. Indicated temperatures were obtained by energy balance calculation (Microsoft EXCEL). Input parameters used for the SOFC-biogas system are summarised in Table 8.

3.3. Energy balance model

Thermal power loss of the isolated module to the surroundings was estimated to 85 W by convection and 33 W by radiation [25]. The sum of 118 W equals 1.3% of the fuel input.

The biogas feed is available at 20-25 mbar above ambient pressure. Total pressure drop for the anode side was calculated to 50 mbar (36 mbar in the reformer, 12 mbar in the

Table 8 Input parameters for the considered SOFC-biogas system

Biogas production (m ³ per day)	35
CH ₄ flow (mol/s)	0.01 (0.25 l/s)
CO ₂ flow (mol/s)	0.00667 (0.167 l/s)
Air flow added to fuel (mol/s)	0.00349 O ₂ (0.417 l/s)
Power equivalent (kW)	8 (LHV), 8.9 (HHV)
Charge equivalent (F)	6.6 (8 F \times 0.99/1.2)
Stack temperature (°C) Fuel utilisation (u_f) (%) Cathode flow $(\lambda = 3)$ (mol/s) Electrolyte resistance $(\Omega \text{ cm}^2)$ Anode resistance $(\Omega \text{ cm}^2)$ Cathode resistance $(\Omega \text{ cm}^2)$ Interconnect contact resistances $(\Omega \text{ cm}^2)$ Total repeat unit resistance $(\Omega \text{ cm}^2)$	800 80 0.2367 (5.61/s) 0.1 0.25 0.1 0.6
Number of cells	100
Stack weight (kg)	7.40
Repeat element height (mm)	3
Stack height (cm)	30
Stack volume (l)	3.4
Stack current (A) Current density (A/cm ²) Cell loss (V)	50.9 (6.6 F C/mol × 0.01 mol/s × $u_{\rm f}$ /100 cells) 0.509 0.305 (0.509 A/cm ² × 0.6 Ω cm ²)
Cell voltage (V)	0.675
Stack voltage (V)	67.5
Stack power (W)	3437 (67.5 V × 50.9 A)
Electrical efficiency (%)	42.8 (LHV), 38.6 (HHV)
Power density (cell) (W/cm ²)	0.344
Power density (volume) (kW/l)	1.01
Power density (weight) (kW/kg)	0.465

stack). Total cathode pressure drop was calculated to 42 mbar (27 mbar in the stack, 14 mbar in the feed circuitry) [25].

The total drop for cathode and anode flow circulation adds up to about 100 mbar. This is in reality reduced because of a backpressure draught ("chimney effect") caused by the hot ascending exhaust gases. Required blowing to compensate for 2500 Pa drop at the anode side $(0.83 \times 10^{-3} \text{ m}^3/\text{s})$ and 5000 Pa drop at the cathode side $(5.6 \times 10^{-3} \text{ m}^3/\text{s})$ gives a parasitic pumping power of 30 W, which is below <1% of the generated electrical power.

The system is divided into a number of subsystems, each of which is treated on an energy balance calculation. The balance for an open system (with exchange of mass) in steady state (no time-dependent terms) is simplified to

$$\sum_{k} \dot{E}_{k}^{+} + \sum_{i} \dot{Q}_{i}^{+} + \sum_{n} \dot{W}_{n}^{+} = 0$$
⁽¹⁾

where the dotted symbols carry the dimension of energy per unit of time and thus represent power (J/s or W) instead of energy (J), where the "+" signs represent an energy gain within the system boundary (energy liberated by the considered system will then represent a negative term on the left hand side of the equation, or a positive term on its right hand side, where the "+" superscript transforms to a "-" superscript), and where "E" represents work ("noble energy" like mechanical or electrical work), "Q" represents heat, and "W" represents the enthalpy of mass components entering and exiting the system boundary.

For a system at constant pressure, "W" corresponds to the specific enthalpies h (J/mol or J/kg) of these components (CH₄, H₂O, CO₂, CO, H₂, O₂, N₂), each multiplied by their mass flow \dot{m} (mole/s or kg/s).

In this system, no heat sources at constant temperature are present that cannot be expressed by mass-energy "W". At most only a single work term is present (the electricity generated by the SOFC stack, \dot{E}_{el}^{-} .) Then Eq. (1) reduces to

$$\sum \dot{m}h_{\rm in} - \sum \dot{m}h_{\rm out} = \dot{E}_{\rm el}^{-} \tag{2a}$$

$$\sum \dot{m}h_{\rm in} - \sum \dot{m}h_{\rm out} = 0 \tag{2b}$$

The enthalpy h of a mass component at a temperature T is calculated as the sum of its standard enthalpy of formation, $h_{\rm f}^{\circ}$, at the reference state (298 K), and its surenthalpy, \hat{h} ,

$$h = h_{\rm f}^\circ + \hat{h}$$

which is defined as

$$\hat{h} = \int_{298\,\mathrm{K}}^{T} C_{\mathrm{p}}\,\mathrm{d}T$$

with C_p the temperature-dependent specific heat capacity (J/kg K or J/mol K) of that component.

The entropy terms are correspondingly defined as $s = s^{\circ} + \hat{s}$, where s° equals the entropy of the component at the reference state (T = 298 K), and the surentropy \hat{s} is

$$\hat{s} = \int_{298\,\mathrm{K}}^{T} \frac{C_p}{T} \,\mathrm{d}T.$$

The specific heat capacities, within the temperature range 273-2000 K are defined by a temperature-dependent function of the type (with *a*, *b*, *c* tabulated constants)

$$a+b\ T+\frac{c}{T^2}.$$

Straightforward integration of this function allows then to compute the surenthalpy and surentropy at any temperature for each of the involved components.

It is finally remarked that HHV = $LHV + mH_2Oq_{cond}$ with $q_{cond} = 44040 \text{ J/mol}$ at 298 K.

With these equations and the input parameters, the SOFC-biogas system in its whole and each of its subsystems can entirely be assessed on its energy balance. The exergy balance was equally performed [25].

3.4. System overview

An overview of the energy balance is given in Fig. 7.

The total input power corresponds to the fuel feed, in the present case 8.024 kW (LHV) or 8.91 kW (HHV) of methane



Fig. 7. Schematic overview of energy balance in the SOFC-biogas system.

(0.01 mol/s). Through the reforming process, which remains overall endothermic (CO_2 -reforming is heavily endothermic, partial oxidation is only slightly exothermic), the fuel calorific power is actually upgraded (to 8.66 kW).

Of this amount, the fraction u_f (80%) is liberated on the SOFC stack (6.92 kW), while the remainder $(1 - u_f)$, or 20%) is generated in the afterburner zone (1.74 kW). The energy liberated on the stack consists of the entropic loss $(u_f T \Delta S)$ due to the chemical reaction (1.93 kW), the balance 4.99 kW being available as theoretically maximal available work $(u_f \Delta G = u_f nFV_G)$, nF being the number of released electrons per mole of fuel, here 6.6 F). Because of voltage drop due to the internal cell resistance (Joule heat), this maximal available work finally splits up in the real achieved work 3.44 kW (electrical output power, $u_f nFV_{cell}$) and further thermal loss 1.55 kW ($u_f nF(V_G - V_{cell})$). This loss together with the entropic loss constitute the heating power generated on the stack (3.48 kW).

This stack heat pre-heats the incoming fuel feed (1.1 kW)and drives the reforming reaction (0.66 kW). The air is pre-heated first by the hot exhaust (to 400–500 °C), then by the afterburner zone heat (to 700 °C) and finally by remaining stack heat to operating temperature, 800 °C. The cooled exhaust (to 500–600 °C) will, upon further cooling, heat the domestic water boiler and compensate for the small system thermal losses.

3.5. Subsystem calculations

The fuel pre-heating process requires 1094 W using Eq. (2b).

From equilibrium calculation at 800 °C the reforming reaction proceeds as:

$$\begin{array}{l} 0.6 \mathrm{CH}_4 + 0.4 \mathrm{CO}_2 + 0.2093 \mathrm{O}_2 \\ \\ \rightarrow 0.0106 \mathrm{CH}_4 + 0.125 \mathrm{H}_2 \mathrm{O} + 0.1056 \mathrm{CO}_2 \\ \\ + 0.8838 \mathrm{CO} + 1.0538 \mathrm{H}_2. \end{array}$$

The power required to drive this endothermic process is calculated from $\sum \dot{m}(h_{\text{out}} - h_{\text{in}})$ to 660 W.

When considering the reformer zone as adiabatic, the theoretical temperature drop that would result from this endothermic reaction can be calculated by applying enthalpy conservation from reactants to products, or

$$H_{\text{reac}} = \sum_{\text{reac}} m_{\text{reac}} \left[h_{\text{f}}^{\circ} + \int_{298}^{1073} C_{p,\text{reac}} \, \mathrm{d}T \right]$$
$$= H_{\text{prod}} = \sum_{\text{prod}} m_{\text{prod}} \left[h_{\text{f}}^{\circ} + \int_{298}^{T_{\text{adiab}}} C_{p,\text{prod}} \, \mathrm{d}T \right]$$

Numerical solution yields $387 \,^{\circ}$ C as result for the unknown temperature T_{adiab} . The reforming reaction without heat exchange would thus cause a 413 K drop in temperature. This indicates the benefit of endothermal reforming (stack heat removal) which reduces the air-fuel ratio value (λ) used on the cathode side.

Fuel conversion was fixed to 80%. This corresponds to directing an amount of $0.792 \text{ mol } O_2$ from the cathode side to a mole of the anode fuel mix. Based on this input, resulting equilibrium concentrations in the anode product stream can

be calculated to give the total electrochemical reaction on the SOFC stack (see Fig. 4):

$$\begin{array}{l} 0.0106\text{CH}_4 + 0.125\text{H}_2\text{O} + 0.1056\text{CO}_2 \\ + 0.8838\text{CO} + 1.0538\text{H}_2 + 0.792\text{O}_2 \\ \rightarrow 0.9833\text{H}_2\text{O} + 0.8204\text{CO}_2 + 0.1796\text{CO} + 0.2167\text{H}_2 \end{array}$$

$$(3)$$

The liberated power on the stack then corresponds to the enthalpy difference between output and input, calculated to 6923 W. Burning 0.01 mol/s of CH₄ at 800 °C liberates 8006 W ($\Delta H_{\text{LHV},800 \circ \text{C},\text{CH}_4} = 800.6 \text{ kJ/mol}$). The reforming reaction requiring an input of 660 W, the fuel mix after reforming has been upgraded to a heating power of 8666 W. Eighty percent of this power, corresponding to the fuel conversion fraction, is liberated on the stack, or 8666 × 0.8 = 6933 W (the difference, 0.14%, being due to rounding errors).

Data consistency is checked by performing the separate calculation for the Gibbs free enthalpy and the various losses involved, such as the chemical reaction entropy and the Joule heating power.

The chemical reaction entropy at 800 °C (for Eq. (3)) was computed to 1932 W. This fraction of fuel energy is not convertible to (electrical) work and generates heat loss. The Gibbs reaction enthalpy (for Eq. (3)) was computed to 4991 W. It is verified that $\Delta G = \Delta H - T\Delta S$, namely, -4991 = -6923 - (-1932).

The value 4991 W corresponds to the maximally obtainable work on the stack. Irreversible loss is calculated as Rl^2 (1554 W) with $R = 0.6 \,\Omega \,\mathrm{cm}^2$ and $I = 50.9 \,\mathrm{A}$ (Table 8). Effective electrical power is then 4991 – 1554 = 3437 W_{el}. From this value, the stack voltage 3437 W/50.9 A = 67.5 V is obtained, the individual average cell voltage U_{cell} (100 cells) is then 0.675 V. The useful power (and thus the electrical efficiency) can only be increased by lowering the repeat unit equivalent resistance (0.6 $\Omega \,\mathrm{cm}^2$), i.e. by improving the cell materials, the interconnect contact and the interface catalysis.

Open circuit voltage (OCV) can be calculated backwards from $U_{cell} = OCV - R I$, or OCV = 0.980 V. Relating this voltage to the Gibbs enthalpy is by considering that:

- 1. 4991 W corresponds to 80% of the available ΔG , hence $\Delta G = -4991/0.8 = -6239$ W for 0.01 mol methane feed, or -623900 W for 1 mol feed;
- 2. the exchanged amount of electrons per oxidized mole of the given biogas-air fuel mix corresponds not to 8 F (as for pure CH₄) but 6.6 F (Table 8) since a portion of the methane was already partially oxidised (to CO) with the added air, prior to conversion into electrical current on the stack. This portion was 0.21/1.20 = 0.175. Hence, $0.175 \times 8 F = 1.4 F$ charge equivalent is lost in the partial oxidation reaction for conversion to electrical current. Therefore, an inherent electrical efficiency drop of 17.5%

with respect to pure biogas feed (without air addition) is present in the system.

With these two considerations, the OCV of $-\Delta G/nF$ or 623900/6.6F = 980 mV is reproduced (see also Fig. 7).

The stack heating power of 3486 W (1932 entropy heat, 1554 W Joule heat) provides power necessary to pre-heat the fuel (1094 W), drive the reformer (660 W) and pre-heat the cathode air flow from its temperature reached in the afterburner zone up to final operating temperature $(800^{\circ},$ Fig. 6). Excess heating power will raise the temperature of the exhaust gases. Based on the experimental observation that exhaust gases reach at the outlet a temperature 100 K higher (i.e. 900 °C) than the average stack temperature, the power transferred to this exhaust from the excess stack heat is computed to 956 W using Eq. (2b). The balance from the stack heating power (i.e. 776 W) will then pre-heat the incoming cathode air, after exchanging with the afterburner zone, up to stack temperature. The entry temperature of the cathode air flow ($\lambda = 3$) to the stack is then computed backwards using Eq. (2b) to yield 700 °C (Fig. 6).

These simplified balance calculations allow to note the importance of the air-fuel ratio λ . If this were doubled from three to six (as would be necessary for cooling a stack fueled on pure hydrogen, for example), the double air mass would have to be pre-heated in this final stage, thus requiring the double heating power Q to be exchanged (1.55 kW in the current example). Simultaneously, this double air mass flow passing the cathodes would have a cooling effect on the outlet stream, which would no more be heated from 800 to 900° but to around half that (850 °C). Therefore, to achieve the same heat transfer with this new situation (λ doubled, Q doubled and ΔT halved), a four-fold higher exchange surface A (i.e. between the cells in the stack) would be necessary, imposing important system design constraints.

Excess stack heat has raised the exhaust gas temperature to 900 °C. In the anode exhaust, 20% unspent fuel is left. This can at the cell rims freely combust with excess air from the cathode outlet stream. This generates another 1744 W of heating power, used to pre-heat incoming cathode air up to 700 °C. Energy balancing (Eq. (2b)) allows to compute backwards that the inlet temperature of this air, exchanging with the hot afterburner zone, equals 470 °C (Fig. 6). The adiabatic temperature rise in the afterburner is computed to 182 K, so exhaust gases would reach, without heat exchange, 1082 °C in the post-combustion zone.

The necessary power to heat the cathode air mass in the first pre-heated portion (Fig. 6), from ambient 15 to 470 °C, is calculated to 3282 W using Eq. (2b). This power is provided by the hottest exhaust, leaving the afterburner at 900°; with Eq. (2b), this exhaust is calculated to cool down to 547 °C in this heat exchange process (Fig. 6).

This final exhaust is available to heat water in a boiler for domestic use, before being rejected to the surroundings. Recovering condensation heat from the water vapour present in the exhaust stream is not considered; the exhaust is rejected

Table 9 SOFC-biogas system: energy balance overview

Fuel feed (biogas/air: 1/1) (mol/s)	0.01 CH ₄ +0.0067 mol/s CO ₂	
System inlet temperature (°C)	15	
System outlet temperature (°C)	60	
Oxidant (air)	$\lambda = 3$	
Fuel inlet power (W)	8024 LHV (8913 HHV)	100%
Produced total power (out-in) (W)	-7659	85.90%
Exhaust loss (W)	-366 LHV (-1254 HHV)	14.10%
SOFC stack temperature (°C)	800	
Fuel conversion (%)	80	
Fuel pre-heater consumption (W)	1094	
Reformer consumption (W)	660 (adiabatic drop to 387 °C)	
SOFC stack power production (W)	-6923 (adiabatic rise to 1505 °C)	
Theoretical maximum work (W)	-4991	
Electrical power (W)	-3437	38.2%
Ventilation needs (W)	30	0.33%
Net electrical power (W)	-3407	
Stack heating power (W)	-3486	
	=-(1094 + 660 + 776 + 956) consumed	
	=(-1932 - 1554) generated	
Entropy heating power (W)	-1932	
Joule heating power (W)	-1554	
Off-gas heatup (800–900 °C) (W)	956	
Air pre-heater III (700-800 °C) (W)	776	
Afterburner (W)	-1744 (adiabatic rise to 1082 °C)	
Air pre-heater II (470–700 °C) (W)	1744	
Air pre-heater I (15–470 $^{\circ}$ C) (W)	3282	
Total air pre-heating power (W)	5979	
Exhaust cooling I (900-547 °C) (W)	-3282	
Exhaust cooling II (547-60 °C) (W)	-4215	
Thermal loss (W)	117	1.31%
Hot water boiler (W)	4098	45.98%

at a temperature close above the highest possible dew point. The highest possible partial pressure of water vapour in the product stream is obtained from the overall reaction

$$\begin{array}{l} (0.6\text{CH}_4 + 0.4\text{CO}_2 + 0.2093\text{O}_2 + 0.79\text{N}_2) \\ + \lambda 0.9907(\text{O}_2 + 3.773\text{N}_2) \\ \rightarrow 1.2\text{H}_2\text{O} + 1\text{CO}_2 + (\lambda - 1)0.9907\text{O}_2 \\ + (0.79 + 3.773\lambda)\text{N}_2. \end{array}$$

Taking the extreme case where $\lambda = 1$, the water vapour content in the product stream will be 18 kPa. With the empirical vapour pressure function

$$p_{\text{sat}}$$
 (Pa) = exp $\left(23.569 - \frac{4041.6}{T_{\text{dew}}$ (°C) + 235.6}\right)

the dew temperature is calculated to 58 °C. For the standard case where $\lambda = 3$, the vapour partial pressure will be only 7.5 kPa and the dew point 40 °C. The exhaust exit temperature is set to 60 °C (Fig. 6). The power contained in the exhaust stream cooled from 547 to 60 °C equals 4215 W using Eq. (2b). For total balancing purposes, the convection and radiation losses of the system are subtracted (117 W), to leave 4.1 kW for domestic hot water use. Eventually lost to the environment is the energy contained in the 60 °C warm

exhaust and the condensation heat of the water vapour. These two figures are 366 (warm exhaust) and 889 W (condensation heat), adding up to 1254 W lost in the chimney (14% of input).

The balance from all subsystems and the data consistency is summarised in Table 9.

4. Conclusions

Sources for biogas generation were reviewed, as well as some European energy policy aspects related to targets of biomass use, CHP installation and emission reductions. Over the period from 1995 to 2010, the use of renewable sources is meant to double, bioelectricity to raise 10-fold and CHP electricity to grow by a factor of 2.5 whereas CO_2 emissions are aimed to decrease by 8% (or 800 mio t per year). Exploitation of biogas potential by SOFCs in small CHP with high electrical conversion, especially on farms (10 kW_{el}), sewage plants (100 kW_{el}) and landfill sites (1 MW_{el}) can significantly contribute to the realisation of these targets.

The current EU-15 biogas production was estimated to 2 Mtoe (world: 25 Mtoe), just 0.13% of primary energy. Its mobilisable potential of 17 Mtoe could generate, by full exploitation with SOFCs, a yearly production of 83 TWh_{el} and

A realistic example of a small SOFC-biogas CHP system (100 cell, 3.41 stack, 8 kW methane LHV) was studied and modelled in terms of an energy balance.

Thermal losses of the module and parasitic electrical losses (for gas circulation) were estimated to be low, 117 W_{th} and 30 W_{el} , respectively. The CO₂ content of biogas does not allow for direct CO₂-reforming. Oxygen (from air) was added here to accomplish the methane reforming to syngas, without risk of carbon desposition above 750 °C. The required oxygen addition was calculated such that biogas and air can be mixed in equal volume flows. The stack delivered 3.4 kW_{el} and 4.1 kW_{th} corresponding to 38.2 electrical and 46% thermal efficiencies, or 84.2% total (HHV), in agreement with the conversion values assumed above.

The heating power generated on the stack (3.5 kW) can pre-heat the fresh fuel (1.1 kW), drive the reformer (0.66 kW) and heat the fresh air to final operating temperature. All fresh air requires 6 kW for a complete heat-up (from ambient to operating temperature), acquired in three phases: from the hot $(900 \degree \text{C})$ exhaust gas (3.3 kW), from ambient to $470 \degree \text{C}$), from the afterburner (1.74 kW), from $470 \text{ to } 700 \degree \text{C}$) and finally from the stack itself (from 700 to $800 \degree \text{C}$). The cooled exhaust is hot enough $(547 \degree \text{C})$ to supply 4.1 kW to a domestic water boiler $(50 \degree \text{C})$, and is rejected at $60 \degree \text{C}$.

The theoretical adiabatic temperature excursions were calculated to be -413 K in the reformer, +705 K in the electrochemical reaction and +182 K in the afterburner.

For future work with this model, it is planned to vary different design and operating parameters (fuel utilisation, temperature, λ , cell geometry and stacking, etc.) to test their influences and obtain the output variations. To this purpose the model is now implemented into an energy systems optimisation tool (BELSIM) [22]. This system study will be applied to different fuel technologies (PEFC/PAFC/MCFC/SOFC) to investigate whether superior performance of the SOFC type is confirmed.

References

- Energy for the Future: Renewable sources of Energy, White Paper for a Community Strategy and Action Plan, COM(97)599 final (26/11/97), Annex1.
- [2] K. Kendall, J. Staniforth, J. Power Sources 71 (1998) 275;
 J. Staniforth, K. Kendall, J. Power Sources 86 (2000) 401–403.
- [3] M. Jenne, T. Dörk, A. Schuler, in: U. Bossel (Ed.), Proceedings of the Fifth European Solid Oxide Fuel Cell Forum, Lucerne, Switzerland, July 2002, European Forum Secretariat, CH 5442-Oberrohrdorf, Switzerland, pp. 460–466.

- [4] Y. Membrez, Biogaz à partir de déchets industriels et ménagers, Publication 724.230f for Office Fédérale des Questions Conjoncturelles, 3003 Bern, Switzerland, 1993.
- [5] O. Bucheli, M. Michielis, Crossing the chasm for Solid Oxide Fuel Cells, Diploma Thesis in Management of Technology (MoT), Secretariat of Postgraduate Programme in MoT, Swiss Federal Institute of Technology Lausanne (EPFL), 1015 Lausanne, Switzerland, 2001.
- [6] Y. Membrez, M. Glauser, Batch anaerobic digestion of agricultural and municipal solid wastes, Publication of Environmental and Energy Services (SEVEN) of Canton Vaud, Switzerland, 1995.
- [7] Nebenerwerb durch Abfallverwertung, Brochure M49d, 5/97, IN-FOENERGIE Suisse Nord-Ouest, c/o Nova Energie GmbH, 5000 Aarau, Switzerland.
- [8] Sewage Plant Exploitation, CH-1100 Morges, Switzerland, private communication.
- [9] Valorisation économique des déchets verts, Brochure M68f, 10/98, INFOENERGIE Suisse Nord-Ouest, c/o Nova Energie GmbH, 5000 Aarau, Switzerland.
- [10] Utilisation énergétique de déchets verts, Brochure M4f, 9/98, IN-FOENERGIE Suisse Nord-Ouest, c/o Nova Energie GmbH, 5000 Aarau, Switzerland.
- [11] H. van den Bergh, Pollution atmosphérique, Postgraduate Course Energy Technology, Swiss Federal Institute of Technology, EPFL, 1998–2000, vol. B.7.1, Secretariat of the Laboratory for Energy Systems.
- [12] N. Houlmann, Gaz naturel, Postgraduate Course Energy Technology, Swiss Federal Institute of Technology, EPFL, 1998–2000, vol. A.5.3, Secretariat of the Laboratory for Energy Systems.
- [13] H. Harms, Energy and organic pollution, Postgraduate Course Energy Technology, Swiss Federal Institute of Technology, EPFL, 1998–2000, vol. B.7.3, Secretariat of the Laboratory for Energy Systems.
- [14] Community Strategy to Promote Combined Heat and Power (CHP) and to Dismantle Barriers to its Development, COM(97) 514 final (15/10/97).
- [15] U. Kaufmann, Schweizerische Statistik der erneuerbaren Energien 2000, Report for the Swiss Federal Energy Office No. 805.520.4 d, CH-3003 Bern, Switzerland.
- [16] Dr. Andrea Tilche, private communication.
- [17] J.-L. Hersener, U. Meier, Energetisch nutzbares Biomassepotential in der Schweiz sowie Stand der Nutzung in ausgewachlten EU-Staaten und den USA, Swiss Federal Energy Office Report, 3003 Bern, Switzerland, April 1999.
- [18] International Energy Agency Report on Bioenergy: Anaerobic Digestion Activity, 1997.
- [19] Novel SOFC system for co-generation of electricity and heat, ENET News No. 9100265-5 (Energy-Net magazine of the Swiss Federal Energy office), Project no. 500, 1996, pp. 228–237.
- [20] K. Ledjeff-Hey, Brennstoffzellentechnologie, Springer Verlag, 2000, ISBN 3-7880-7629-1.
- [21] C. Rösch, Energetische Nutzung von Biomasse mit Brennstoffzellen, Gülzower Fachgespräche, 1998, pp. 18–25;
 I.C. Vinke, Energetische Nutzung von Biomasse mit Brennstoffzellen, Gülzower Fachgespräche, 1998, pp. 110–113.
- [22] J. Van herle, F. Maréchal, S. Leuenberger, D. Favrat, J. Power Sources 118 (2003) 375–383.
- [23] J. Van herle, Y. Membrez, Biogas exploitation in SOFC, in: U. Bossel (Ed.), Proceedings of the Fifth European Solid Oxide Fuel cell Forum, Lucerne, Switzerland, July 2002, European Forum Secretariat, CH 5442-Oberrohrdorf, Switzerland, pp. 1003–1010.
- [24] J. Van herle, R. Ihringer, R. Vasquez, L. Constantin, O. Bucheli, Anode supported SOFC with screen-printed cathodes, J. Eur. Ceram. Soc. 21 (2001) 1855–1859.
- [25] J. Van herle, SOFCs fueled with biogas, Masters Thesis in Energy Technology, Swiss Federal Institute of Technology, Lausanne, Switzerland, Secretariat of the Laboratory for Energy Systems, 2001.