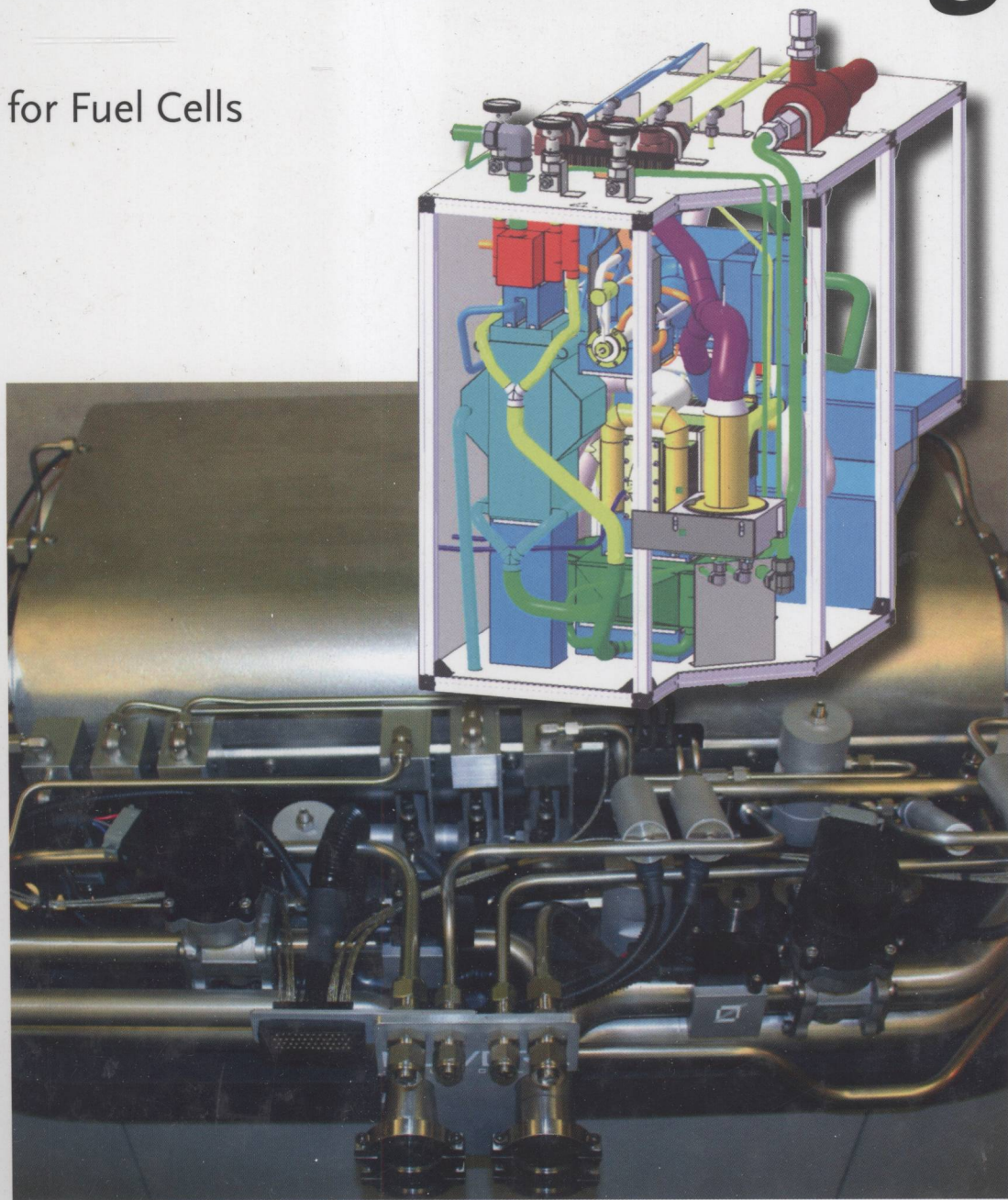


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Further Reading

K. Sundmacher, A. Kienle, H. J. Pesch, J. F. Berndt, G. Huppmann (Eds.)

Molten Carbonate Fuel Cells

Modeling, Analysis, Simulation, and Control

2007

ISBN: 978-3-527-31474-4

W. Vielstich, A. Lamm, H. Gasteiger (Eds.)

Handbook of Fuel Cells - Fundamentals, Technology, Applications

4 volume set

2003

ISBN: 978-0-471-49926-8

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2007

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Gunther Kolb

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1

Introduction and Outline

Mankind's energy demand is increasing exponentially. Between 1900 and 1997, the world's population more than tripled and the average energy demand per human being has also more than tripled, resulting in greater than thirteen times higher overall global emissions [1]. Thus the carbon dioxide concentration rose from 295 parts per million in 1900 to 364 parts per million in 1997 [1]. In 1997 almost all European countries committed to reducing greenhouse gas emissions to an amount 8% below the emissions of 1990 in the period from 2008 to 2012. With this scenario, fuel cell technology is attracting increasing attention nowadays, because it offers the potential to lower these emissions, owing to a potentially superior efficiency compared with combustion engines. Fuel cells require hydrogen for their operation and consequently numerous technologies are under investigation worldwide for the storage of hydrogen, aimed at distribution, and mobile and portable applications.

The lack of a hydrogen infrastructure in the short term, along with the highly attractive energy density of liquid fossil and regenerative fuels, has created widespread research efforts in the field of distribution and on-board hydrogen generation from various fuels. This complex chemical process, generally termed fuel processing, is the subject of this book.

The electrical power output equivalent of the fuel processors that are currently under development world wide covers a wide range, from less than a watt to several megawatts. Portable and small scale mobile fuel cell systems promise to be the first commercial market for fuel cells, according to a market study of *Fuel Cell Today* in July 2003 [2]. According to the same report, the number of systems built has increased dramatically to up to more than 3000 in 2003. To date, most of these systems have used Proton Exchange Membrane (PEM) fuel cells.

Low power fuel processors (1–250 W) compete with both conventional storage equipment, such as batteries, and simpler fuel cell systems, such as Direct Methanol Fuel Cells (DMFC).

Fuel cell systems for residential applications are typically developed for the generation of power and heat, which increases their overall efficiency considerably, because even low temperature off-heat may be utilised for hot water generation, which reduces energy losses considerably.

For mobile applications, systems designed to move a vehicle need to be distinguished from the Auxiliary Power Unit (APU), which either creates extra energy for the vehicle (e.g., the air conditioning and refrigerator system of a truck) or works as a stand alone system for the electrical power supply.

This book provides a general overview of the field of fuel processing for fuel cell applications. Its focus is on mobile, portable and residential applications, but the technology required for the smaller stationary scale is also discussed.

In the second chapter fundamental definitions and the basic knowledge of fuel cell technology are provided, as far as is required to gain an insight into the interplay between the fuel cell and its hydrogen supply unit – the fuel processor.

The third chapter deals with the reforming chemistry of conventional and alternative fuels, and with the chemistry of catalytic carbon monoxide clean-up, sulfur removal and catalytic combustion.

An overview of catalyst technology for fuel processing applications is provided in Chapter 4, covering all the processes described in Chapter 3.

The design of the individual components of the fuel processor is the subject of Chapter 5. Design concepts and numerical simulations presented in the open literature are discussed for reforming, catalytic carbon monoxide clean-up and physical clean-up strategies, such as membrane separation and pressure swing adsorption. In addition, fuel processor concepts are then presented and the interplay between the various fuel processor components is explained. Details of the basic engineering of fuel processors and dynamic simulations are discussed, covering start-up and control strategies. Some tips and the basic knowledge required to perform such calculations are provided.

There are three basic types of fuel processing reactors, namely fixed catalyst beds, monoliths and plate heat-exchangers, which are explained in Chapter 6.

Chapter 7 then shows the practical applications of such reactors, as published in the literature.

In Chapter 8 some important aspects of balance-of-plant components are discussed, and Chapter 9 presents complete fuel processors for all types of fuels, while cost and production issues are the subject of Chapter 10.

2 Fundamentals

This chapter provides information about common fossil fuels, necessary definitions in the field of fuel processing and the basic knowledge from the wide field of fuel cell technology. It is by no means comprehensive and is not a substitute for the dedicated literature in these fields. Rather, it provides a brief summary for readers who wish to gain an overview of the topic of fuel processing without the need to use too much additional literature.

2.1 Common Fossil Fuels

Fuels are solid, liquid or gaseous energy carriers. To date, practically all of the fuels available on the market are based upon fossil sources and thus contain hydrocarbons of varying composition. However, alternative fuels such as alcohols and hydrides may serve as future energy carriers. Table 2.1 provides an overview of the conventional fuels and of the most important alternative fuels, which may act as future hydrogen source for fuel cells along with their key properties.

A comparison of the gravimetric and volumetric density of various hydrogen carriers shows that liquid hydrocarbons have – apart from borohydrides – by far the best combined properties (see Figure 2.1).

Table 2.2 shows the maximum amount of work that can be converted into electricity from various fuels, in theory. Compared with the gravimetric and volumetric energy density of 1 MJ kg^{-1} or $<2 \text{ MJ L}^{-1}$ of lithium-ion and zinc-air batteries, these values are considerably higher.

The composition of fossil hydrocarbon fuels may vary widely depending on the source of the crude oil that is processed in the refinery.

The composition of natural gas is predominantly methane, and also contains several percent ethane and propane. In addition, minor amounts of butane and higher hydrocarbons are present, plus carbon dioxide and nitrogen.

Table 2.3 shows the composition of natural gas from various sources [5]. Natural gas also contains sulfur compounds at the ppm-level, such as hydrogen sulfide and

Table 2.1 Overview of important fuels for fuel processing.

Fuel	Formula	Sulphur content [wt. ppm] [only commercially available fuels]	Lower heating value [kJ/mol]	Flammability limits lower, higher [Vol.%]	Density [kg/m ³]	Boiling point or boiling range [°C]	Heat of vaporization [kJ/mol]	Heat capacity [J/(mol K)] At 20°C
Hydrogen	H ₂	–	240	4.1, 74	0.09	–252.7	0.92	28.6
Methanol	CH ₃ OH	–	643	7.3, 36	794 (l)	64.6	35.2	49.0 (g)
Ethanol	C ₂ H ₅ OH	–	1240	4.3, 19	790 (l)	78.3	38.9	77.3
Methane	CH ₄	–	802	–	0.72	–161.4	8.2	35.5
Natural gas	C _{1.07} H _{4.1}	7–25	797	5.3, 15	0.77	–	8.2	–
Propane	C ₃ H ₈	–	2015	2.2, 9.5	1.96	–42	73.5	73.5
Liquefied Petroleum Gas [LPG]	C ₃ H ₈ /C ₄ H ₁₀	50–200	2024	1.5, 11	540 (l) (at 8 bar)	–42––0.5	–	–
Iso-octane	C ₈ H ₁₈	–	4731	–	–	99.3	34.8	–
Gasoline	C _{7.1} H _{14.3}	150 50 (European Regulation 2005)	4,720	0.8, 8	720–770	30–200	33.5	180
Dodecane	C ₁₂ H ₂₆	–	7,392	–	750	216.4	40.6	270.2
Hexadecane	C ₁₆ H ₃₄	–	9,792	–	770	286.8	51.3	386.5
Diesel	C _{13.6} H _{27.1}	50 (European regulation 2005) Heating oil 2,000	8,080	1, 6	0.856	120–430	47.0	340
Bio-diesel	C _{18.7} H _{34.5} O ₂	<3,000	10,800	–	–	–	67.8	–

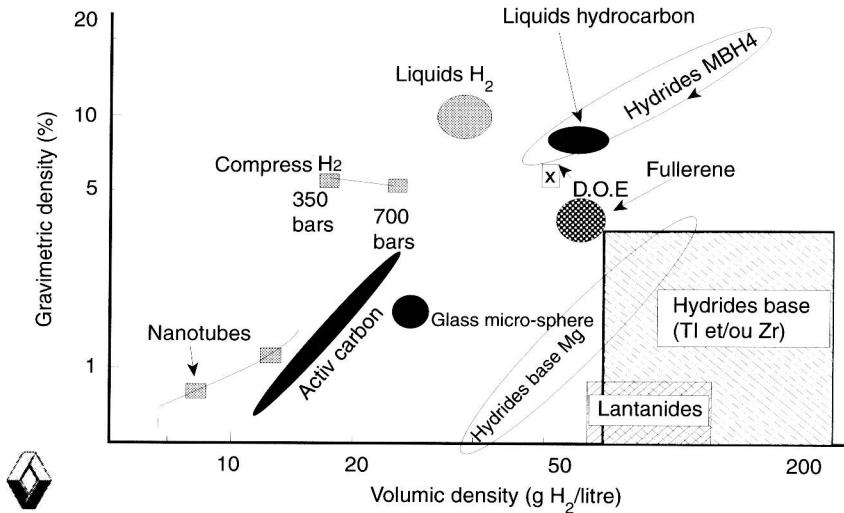


Figure 2.1 Comparison of gravimetric and volumetric storage densities as provided by Heurtaux *et al.* [3].

diethyl sulfide, and mercaptanes, such as ethyl mercaptane [(C₂H₅)CHS] and tertiary butyl mercaptane [(CH₃)₃CHS].

Amongst all the fossil fuels, propane contains the highest amount of hydrogen on a gravimetric basis, which even exceeds liquefied hydrogen, when the weight of the storage tanks is taken into consideration [6]. Propane is usually marketed as liquefied petroleum gas, which is a mixture of propane and butane in various ratios.

For gasoline, only approximate characterization parameters are provided, such as the octane number, the boiling point distribution, and the saturated hydrocarbons (alkanes), unsaturated hydrocarbons (olefins) and aromatics content. The content of contaminants, such as sulfur, is important.

Table 2.2 Energy density of various fuels related to different properties [4].

Fuel	Maximum amount of work				MJ/Mol H ₂ via reforming
	MJ/Mol fuel	MJ/Kg fuel	MJ/L fuel	MJ/Mol C in fuel	
Methanol	-0.69	-22	-17 ^a	-0.69	-0.23
Ethanol	-1.31	-28	-22 ^a	-0.65	-0.22
n-Octane	-5.23	-46	-32 ^a	-0.65	-0.21
Ammonia	-0.33	-19	-10 ^a		-0.22
Methane	-0.80	-50	-3.9 ^b	-0.80	-0.20
Hydrogen	-0.23	-113	-0.89 ^b		-0.23

^a density of the liquid fuels calculated at 298K and 1 bar, for ammonia at 10 bar.

^b density of the gaseous fuels calculated at 298K and 100 bar.

Table 2.3 Composition of natural gas from various sources [5].

Component	North Sea	Qatar	Netherlands	Pakistan	Ekofisk
CH ₄ (Vol.%)	94.86	76.6	81.4	93.48	85.5
C ₂ H ₆ (Vol.%)	3.90	12.59	2.9	0.24	8.36
C ₃ H ₈ (Vol.%)		2.38	0.4	0.24	2.85
<i>i</i> -C ₄ H ₁₀ (Vol.%)	0.15	0.11		0.04	0.86
<i>n</i> -C ₄ H ₁₀ (Vol.%)		0.21	0.1	0.06	
C ₅ ⁺ (Vol.%)		0.02		0.41	0.22
N ₂ (Vol.%)	0.79	0.24	14.2	4.02	0.43
S (ppm)	4	1.02	1	N/A	30

Regular gasoline, at least according to German standards, is well represented by the overall formula C₇H₁₂ [7].

A standard jet fuel that is frequently cited is the American JP-8 fuel. It contains about 1 000 ppm sulfur and up to 1.5 vol.% non-volatile hydrocarbons [8, 9]. Jet fuels widely used in the world are Jet fuel A and A1 [10] with a boiling range between 150 and 300 °C.

Diesel fuels contain mainly iso-paraffins, but also *n*-paraffins, mono-, di-, tri-, tetra-cycloparaffins, alkylbenzenes, naphthalenes and phenanthrenes and even pyrenes [11].

2.2

Basic Definitions, Calculations and Legislation

Fuel processing is the conversion of hydrocarbons, alcohol fuels and other alternative energy carriers into hydrogen containing gas mixtures. The chemical conversion is achieved in most instances in the gaseous phase, normally heterogeneously catalysed in the presence of a solid catalyst and less frequently homogeneously at high temperature without a catalyst.

The first step of the conversion procedure is generally termed reforming, and has been well established in large scale industrial processes for many decades. The industrial applications most commonly (about 76% [12]) use natural gas as feedstock. The purpose of this process is the production of synthesis gas, a mixture of carbon monoxide and hydrogen, which is then used for numerous processes in large scale chemical production, which are not subject of this book.

Rather, the focus of this book is the technology that provides a hydrogen containing gas mixture, termed the reformat, which is suitable for feeding into a fuel cell. The fuel cell then converts hydrogen into electrical energy. Carbon monoxide may also be converted, which depends on the fuel cell type (see Section 2.3.2).

The lower heating value of a chemical substance is defined as its standard enthalpy of formation. The lower heating value of any fuel C_xH_yO_z is easily determined by the following formula [13]:

$$LHV[\text{kJ mol}^{-1}] = \left(\frac{y}{2} + 2x - z\right)198.8 + 25.4 \quad (2.1)$$

The performance of a fuel processor is measured by its overall efficiency, which is commonly defined as the ratio between the Lower Heating Value (LHV) of the hydrogen and carbon monoxide that are produced to the LHV of the fuel consumed:

$$\eta_{\text{Fuel processor}} = \frac{LHV(\text{H}_2) n_{\text{H}_2} + LHV(\text{CO}) n_{\text{CO}}}{LHV(\text{Fuel}) n_{\text{Fuel}}} \quad (2.2)$$

n are the molar flows and the lower heating value is in units of kJ mol^{-1} . The efficiency of the reformer may be calculated by a simplified version of Eq. (2.2):

$$\eta_{\text{Reformer}} = \frac{LHV(\text{H}_2) n_{\text{H}_2}}{LHV(\text{Fuel}) n_{\text{Fuel}}} \quad (2.3)$$

A certain portion of the hydrogen produced by the fuel processor is frequently fed back to it, because it is not completely consumed by the fuel cell (see Section 2.3). The curious situation may then arise where the fuel processor efficiency exceeds 100%. In particular, this is the situation for steam reforming, where substantially more heat is required to run the process compared with partial oxidation and autothermal reforming (see Section 3). A fuel processor running on steam reforming may reach up to 120% efficiency according to the Eqs. (2.2) and (2.3).

The carbon monoxide content of the reformat obviously needs to be minimised for low temperature proton exchange membrane fuel cells, but other fuel cells may well utilize it as a fuel (see Section 2.3.2). The same applies for methane in certain fuel cells. Therefore, the heating value of the hydrogen alone does not provide the appropriate number for the calculation of efficiency in this instance.

A modified definition of the fuel processor efficiency provides a more realistic value than Eqs. (2.2) and (2.3) [14]:

$$\eta_{\text{Fuel processor}} = \frac{LHV(\text{H}_2) n_{\text{H}_2} + LHV(\text{CO}) n_{\text{CO}} + LHV(\text{CH}_4) n_{\text{CH}_4} - [LHV(\text{H}_2) n_{\text{H}_2} + LHV(\text{CO}) n_{\text{CO}} + LHV(\text{CH}_4) n_{\text{CH}_4}]_{\text{recirculated}}}{LHV(\text{Fuel}) n_{\text{Fuel}}} \quad (2.4)$$

In addition to the formula provided by Lutz *et al.* [14], it takes into consideration the release of unconverted methane and the formation of methane by the reforming process (see Section 3). Unconverted methane is commonly re-circulated to the fuel processor, along with unconverted carbon monoxide, in particular for high temperature fuel cells.

However, for PEM fuel cells methane and carbon monoxide could be excluded from efficiency calculations, because they are not converted in the fuel cell.

The following definition of efficiency was proposed by Feitelberg [15]. It was modified to also take methane and carbon monoxide fed to the fuel cell into consideration as discussed above:

$$\eta_{\text{Fuel processor}} = \frac{LHV(\text{H}_2) n_{\text{H}_2} + LHV(\text{CO}) n_{\text{CO}} + LHV(\text{CH}_4) n_{\text{CH}_4}}{LHV(\text{Fuel}) n_{\text{Fuel}} + [LHV(\text{H}_2) n_{\text{H}_2} + LHV(\text{CO}) n_{\text{CO}} + LHV(\text{CH}_4) n_{\text{CH}_4}]_{\text{recirculated}}} \quad (2.5)$$