

High Energy Batteries

C.-H. DUSTMANN

10.1 INTRODUCTION

As I write this in the year 2002 electric vehicles (EVs) are practically irrelevant for road transport ([Figure 10.1](#)). In the year 2000 there were 109 electric cars registered in Germany out of 3,378,343 total (0.003%). Why do we talk about EVs at all?

Electricity is widely used in nearly all industrial and private areas because it can be converted easily into heat, light, and motion and runs all electric devices. Electricity is convenient, clean where it is used, and economical. To an increasing extent it is used with batteries in telephones, computers, tools, etc., independent from the direct connection to a power plant.

Electric motors with inverters using modern power electronics have the perfect characteristics for city vehicles. Due to the high torque from zero speed no clutch is necessary. Overload capability for acceleration makes an 18-kW electric motor more dynamic than a 42-kW gasoline engine ([Figure 10.2](#)). Electric vehicles are quiet, have no emissions and offer the option to use any renewable primary energy for mobility.

The only reason for which electric vehicles are used to the very limited extent they are now is the battery—the key component for the performance and autonomy of electric vehicles. In the following chapters those battery systems will be described that offer a specific energy of about 100 Wh/kg. This specific energy is necessary for the minimum range of 100 km in Europe or 100 miles in the United States under all normal driving conditions for a marketable electric vehicle. [Figure 10.3](#) shows a substitution potential of 25% of cars in private households if the vehicle range is

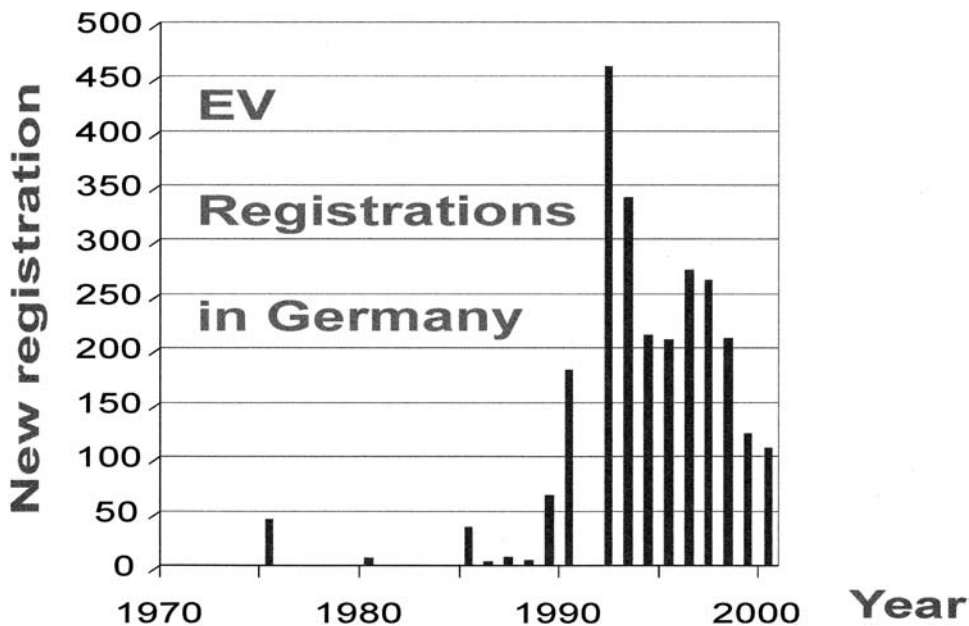


Figure 10.1 30 years EV registrations in Germany. (From Ref. 1.)

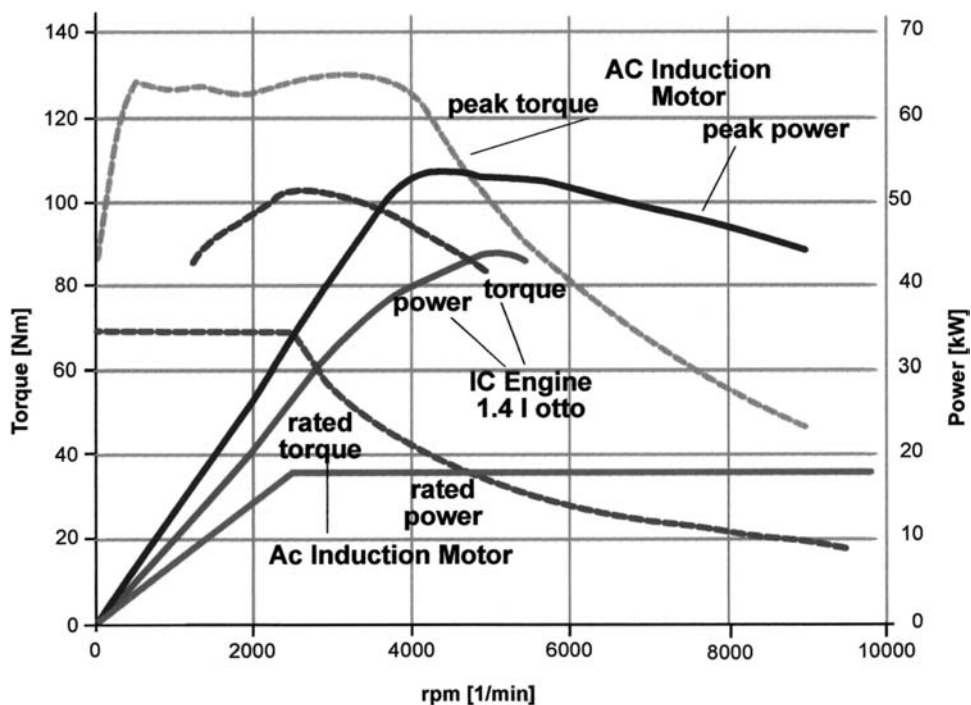


Figure 10.2 Torque and power characteristic of an electric motor with rated power of 18 kW and a 1.4 L gasoline engine. (From Ref. 2.)

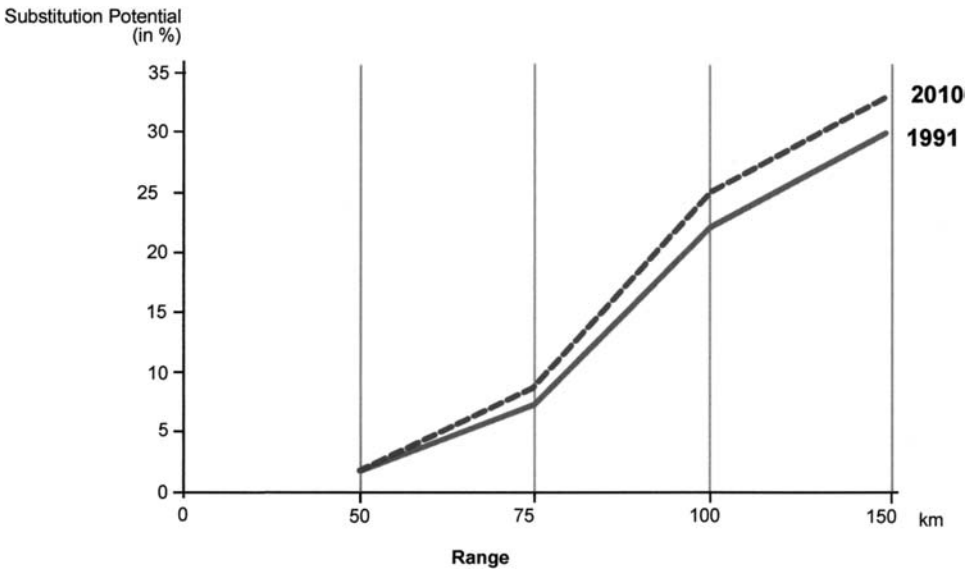


Figure 10.3 Substitution potential of electric vehicles dependent on the vehicle range without change of mobility behavior. (From Ref. 3.)

100 km without a change in the mobility behavior of the users. This is the result of an empirical mobility study [3].

As soon as such a battery is available in sufficient quantities and at a reasonable price, electric vehicles will be available at least for urban transportation. A cost comparison to conventional vehicles is presented in Section 10.7.2. [Table 10.1](#) gives an overview of potential candidates from the present point of view.

10.2 ZEBRA BATTERY (Na/NiCl₂)

10.2.1 Technology

ZEBRA batteries use Ni power and plain salt for the electrode material; the electrolyte and separator is β'' -Al₂O₃-ceramic, which is conductive for Na⁺ ions but an insulator for electrons [4].

This sodium ion conductivity has a reasonable value of $\geq 0.2 \Omega^{-1} \text{cm}^{-1}$ at 260 °C and is temperature dependent with a negative gradient [5]. For this reason the operational temperature of ZEBRA batteries has been chosen in the range of 270 to 350 °C.

[Figure 10.4](#) illustrates the principle. During charge the salt (NaCl) is decomposed to sodium (Na) and chlorine (Cl). The sodium is ionized; one electron from the m3 shell is conducted by the charger to the higher potential of the anode (minus pole), where it recombines with the sodium ion (Na⁺) which was conducted through the β'' -Al₂O₃ electrolyte. The free chlorine reacts with nickel (Ni) in the vicinity to form nickel chlorine (NiCl₂) as a thin layer that covers the nickel grains.

Table 10.1 EV battery systems.

System	Pb/Pbo	NiMH	Na/NiCl ₂	Na/S	Li-ion	LPB
Operating temperature (°C)	<45	<45	235–350	285–330	<50	60–80
Electrolyte	H ₂ SO ₄	KOH	β'' -ceramic	β'' -ceramic	LiPF ₆	Polyethylene oxide
Cell OCV (V)	2.0	1.2	2.58	2.1	4.0	4.0
Specific energy (Wh/kg)	25–35	40–60	100–120	110	80–120	100–120
Energy density (Wh/L)	50–90	120–160	160–200	135	200	200
Specific power (W/kg)	150	Up to 1000	150–180	<75	500–800	300–400
Comments	Largest use	^a	EV battery	Stationary	^b	

^aVery high power cells for power assist HEV are available.

^bLi-ion batteries can be optimized for high power or high energy.

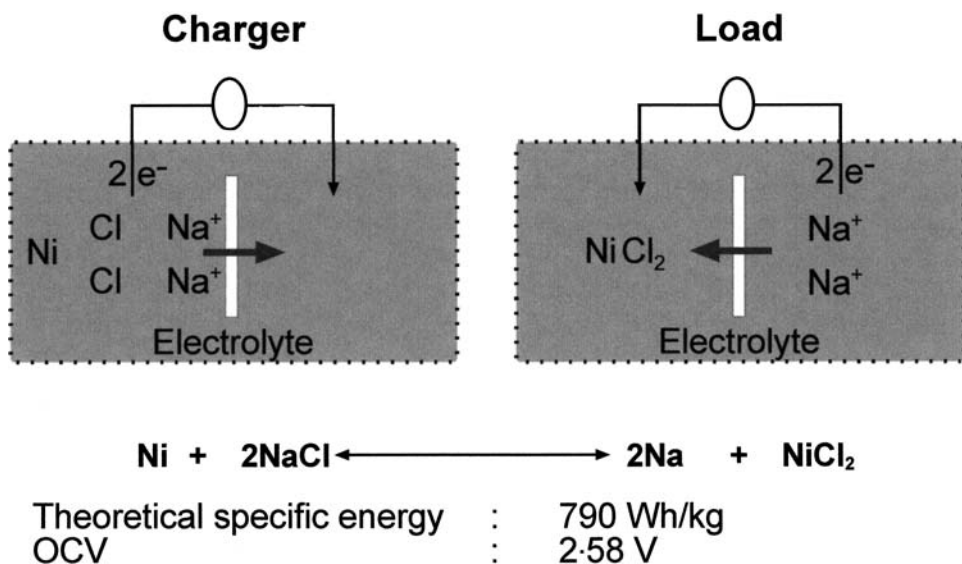


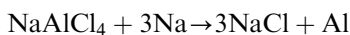
Figure 10.4 ZEBRA chemistry.

The reverse reaction during discharge is only possible by ionization of the sodium; the sodium ion is conducted back through the $\beta''\text{-Al}_2\text{O}_3$ electrolyte to the cathode, whereas the electron now delivers its energy that was previously taken from the charger to the load. In the cathode it recombines with the sodium to form salt and nickel again.

There is no side reaction and therefore the charge and discharge cycle has 100% charge efficiency; no charge is lost. This is due to the ceramic electrolyte.

The cathode has a porous structure of nickel and salt which is impregnated with NaAlCl_4 , a 50/50 mixture of NaCl and AlCl_3 . This salt liquefies at 154°C , and in the liquid state it is conductive for sodium ions. It has the following functions, which are essential for ZEBRA battery technology:

1. Sodium ion conductivity inside the cathode. The ZEBRA cells are produced in the discharged state. The liquid salt NaAlCl_4 is vacuum impregnated into the porous nickel/salt mixture that forms the cathode. It conducts the sodium ions between the $\beta''\text{-Al}_2\text{O}_3$ ceramic surface and the reaction zone inside the cathode bulk during charge and discharge and makes all cathode material available for energy storage. It also provides a homogenous current distribution in the ceramic electrolyte.
2. Low resistive cell failure mode. Ceramic is a brittle material and may have a small crack or may break. In this case the liquid salt NaAlCl_4 gets into contact with the liquid sodium (the melting point of sodium is 90°C) and reacts to salt and aluminum:

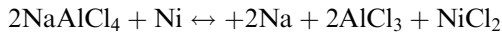


In case of small cracks in the $\beta''\text{-alumina}$ the salt and aluminum closes the crack. In case of a large crack or break the aluminum formed by the above

reaction shorts the current path between plus and minus so that the cell goes to low resistance. By this means long chains of 100 or 200 cells only lose the voltage of one cell (2.58 V) but can continue to be operated. The ZEBRA battery is cell-failure tolerant. It has been established that 5 to 10% of cells may fail before the battery can no longer be used.

This same reaction of the liquid salt and liquid sodium is relevant for the high safety standard of ZEBRA batteries: In case of mechanical damage of the ceramic separator due to a crash of the car the two liquids react in the same way, and the salt and aluminum passivates the NiCl_2 cathode. The energy released is reduced by about 1/3 compared to the normal discharge reaction of sodium with nickel chloride.

3. Overcharge reaction. The charge capacity of the ZEBRA cell is determined by the quantity of salt (NaCl) available in the cathode. In case a cell is fully charged and the charge voltage continues to be applied to the cell for whatever reasons, the liquid salt NaAlCl_4 supplies a sodium reserve following the reversible reaction



This overcharge reaction requires a higher voltage than the normal charge, as illustrated in Figure 10.5. This has three practical very welcome consequences:

OCV vs SOC

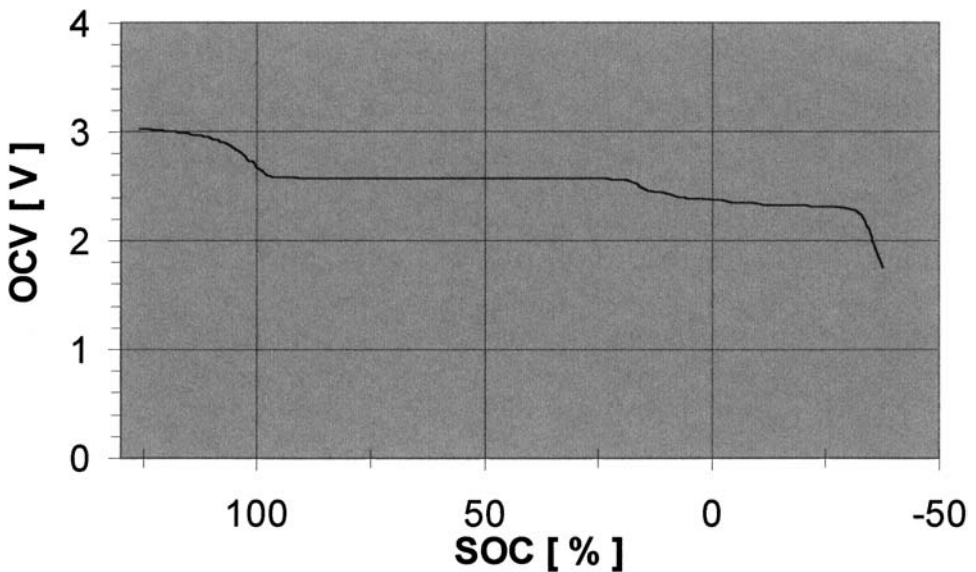


Figure 10.5 ZEBRA open circuit voltage (OCV) depending on the status of charge (SOC).

- (a) Any further charge current is stopped automatically as soon as the increased open voltage equalizes the charger voltage.
 - (b) If cells are failed in parallel strings of cells in a battery, the remaining cells in the string with the failed cells can be overcharged in order to balance the voltage of the failed cells.
 - (c) For a vehicle fully charged in mountainous conditions there is an overcharge capacity of up to 5% for regenerative braking so that the break behavior of the vehicle is fundamentally unchanged.
4. Overdischarge reaction. From the very first charge the cell has a surplus of sodium in the anode compartment so that for an overdischarge tolerance sodium is available to maintain current flow at a lower voltage, as indicated in Figure 10.5. This reaction is equal to the cell failure reaction but runs without a ceramic failure.

10.2.2 ZEBRA Cell Design and Production

ZEBRA cells are produced in the discharged state so that no metallic sodium can be handled. All the required sodium is inserted as salt. Figure 10.6 shows the cell design. The positive pole is connected to the current collector, which is a hair-needle shaped wire with an inside copper core for low resistivity and an outside nickel plating so that all material in contact with the cathode is consistent with the cell chemistry.

The cathode material in form of a granulated mixture of salt with nickel powder and traces of iron and aluminum is filled into the β -alumina tube (Figure 10.7). This tube is corrugated for resistance reduction by the increased surface and is surrounded and supported to the cell case by a 0.1-mm-thick steel sheet that forms a capillary gap surrounding the β -alumina tube. Due to capillary force the sodium is wicked to the top of the tube and wets it independently of the sodium level in the anode compartment.

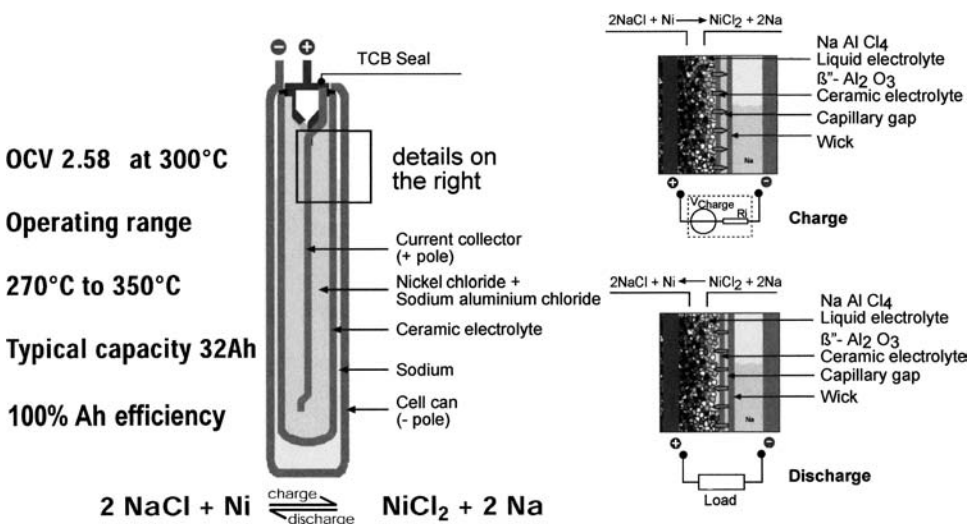


Figure 10.6 Typical ZEBRA cell design.

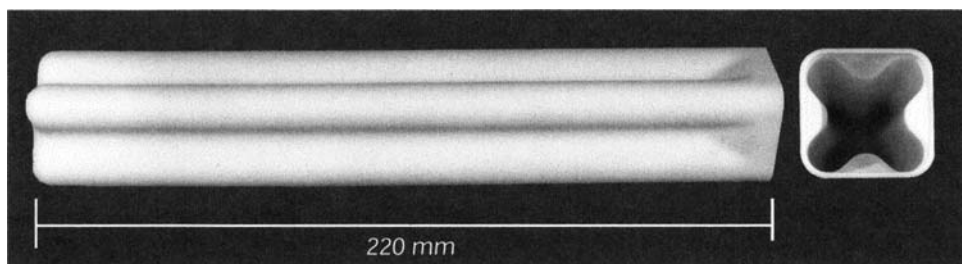


Figure 10.7 Beta-alumina tube.

The cell case is formed out of a rectangular tube continuously welded and formed from a nickel-coated steel strip and a laser-welded bottom cap. The cell case forms the negative pole.

The cell is hermetically sealed by laser-welded nickel rings that are thermocompression bounded (TCB) to an α -alumina collar which is glass brazed to the β -alumina tube.

10.2.3 ZEBRA Battery Design and Production

ZEBRA cells can be connected in parallel and in series. Different battery types have been made with one to five parallel strings, up to 220 cells in series, and 100 to 500 cells in one battery pack. The standard battery type Z5 (Figure 10.8) has 216 cells arranged in one (OCV = 557 V) or two (OCV = 278 V) strings. Between every second cell there is a cooling plate through which ambient air is circulated (Figure 10.9), providing a cooling power of 1.6 to 2 kW. For thermal insulation and mechanical support the cells are surrounded by a double-walled vacuum insulation typically 25 mm thick. Light plates made out of foamed siliconoxide take the atmospheric pressure load. This configuration has a heat conductivity of only 0.006 W/mK and is stable for up to 1000 °C.

Type		Z5-278- ML-64	Z5-557- ML-32
Capacity	Ah	64	32
Rated Energy	kWh	17.8	17.8
Open Circuit Voltage			
0-15% DOD	V	278.6	557
Max. discharge current	A	224	112
Cell Type/N° of cells		ML3 / 216	
Weight with BMI	kg	195	
Specific energy without BMI	Wh/kg	94	
Energy density without BMI	Wh/l	148	
Specific power	W/kg	169	
Power density	W/l	265	
Peak power	kW	32	
80% DOD, 2/3 OCV, 30s, 335°C			
Ambient temperature	°C	-40 to +50	
Thermal loss at 270°C internal temperature	W	< 110	

Figure 10.8 Standard ZEBRA battery type Z5C.



Figure 10.9 Z5C battery cooling plates.

10.2.4 Battery System Design

Figure 10.10 illustrates all components of the complete system ready for assembly. The ohmic heater and the fan for cooling are controlled by the battery management interface (BMI) for thermal management. Plus and minus poles are connected to a main circuit breaker that can disconnect from outside the battery. The circuit breaker is also controlled by the BMI.

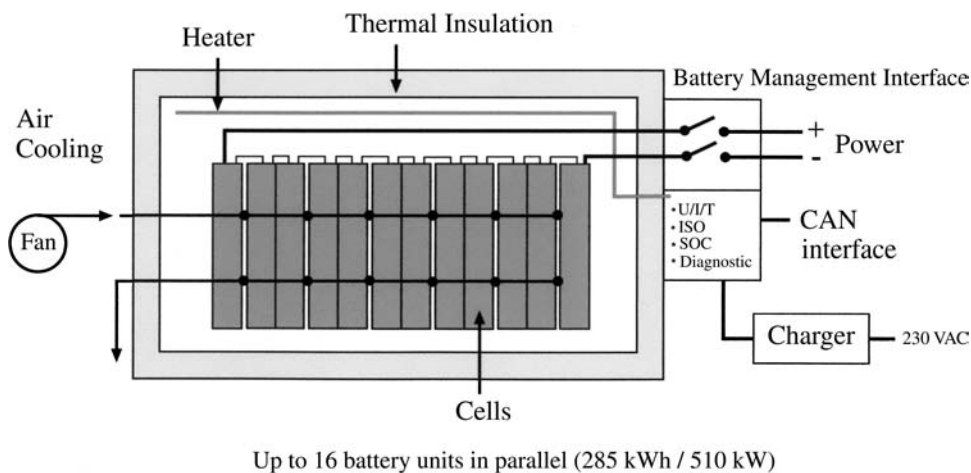


Figure 10.10 ZEBRA battery system.

The BMI measures and supervises voltage, current, status of charge, and insulation resistance of plus and minus to ground and also controls the charger by a dedicated PWM signal. A CAN-bus is used for the communication between the BMI, the vehicle, and the electric drive system. All battery data are available for monitoring and diagnostics with a notebook computer.

A multibattery server is designed for up to 16 battery packs to be connected in parallel in a multibattery system with 285 kWh/510 kW using Z5C batteries.

10.2.5 ZEBRA Battery Performance and Life Data

ZEBRA cells and batteries are charged in an IU characteristic with a 6-h rate for normal charge and a 1-h rate for fast charge. The voltage limitation is 2.67 V/C for normal charge and 2.85 V/C for fast charge. Fast charge is permitted up to 80% SOC. Regenerative braking is limited to 3.1 V/C and 60 A/C so that high regenerative braking rates are possible.

The peak power during discharge, defined as the power at 2/3 OCV, is independent of SOC so that the vehicle performance and dynamic is constant over the whole SOC range [6]. Obviously this is important for practical reasons. Typical battery parameters are summarized in [Figure 10.8](#).

Battery life is specified as calendar life and cycle life. The calendar life of 11 years is demonstrated. The cycle life is measured by the accumulation of all discharged charge measured in Ah divided by the nameplate capacity in Ah, so that one nameplate cycle is equivalent to a 100% discharge cycle. This is a reasonable unit because of the 100% Ah efficiency of the system. Furthermore 100% of the nameplate capacity is available for use without influence on battery life. The expected cycle life is up to 2500 nameplate cycles.

10.2.6 Battery Safety

Battery safety is essential, especially for mobile applications keeping in mind that each battery should store as much energy as possible, but this energy must not be released in an uncontrolled way under any conditions. It is required that even in a major accident there is no additional danger originating from the battery. Many different tests are performed to ensure safety, e.g., crash tests of an operative battery against a pole at 50 km/h ([Figure 10.11](#)), overcharge tests, overdischarge tests, short circuit tests, vibration tests, external fire tests, and submersion tests of the battery in water have been specified and performed [7]. The ZEBRA battery passed all these tests because it employs a four-barrier safety concept [8,9]:

1. Barrier by the chemistry. In case of severe mechanical damage of the battery the brittle ceramic breaks, whereas the cell case made out of steel is deformed and most likely remains closed. In any case the liquid electrolyte reacts with the liquid sodium to form salt and aluminum equal to the overcharge reaction described above. These reaction products form a layer covering the NiCl_2 cathode and thus passivate it. This reaction reduces the thermal load by about 1/3 compared to the total electrochemically stored energy.

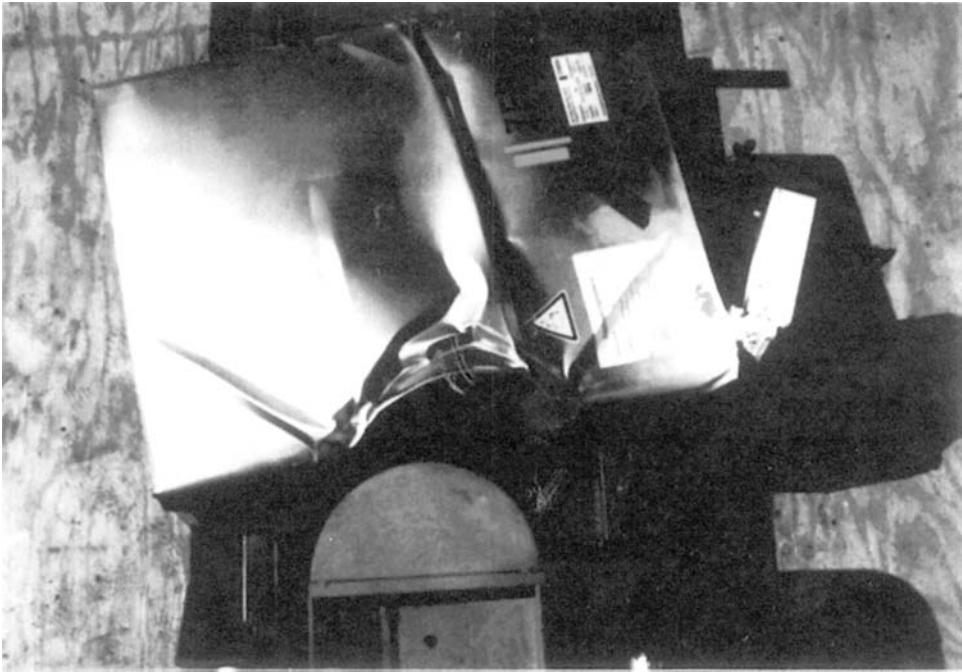


Figure 10.11 ZEBRA battery crashed against a pole at 50 km/h.

2. Barrier by the cell case. The cell case is made out of steel with glass-brazed thermocompression-bounded seal that remains closed for temperatures up to about 900 °C.
3. Barrier by the thermal enclosure. The thermal isolation material of the battery box is made out of foamed SiO_2 which is stable for above 1000 °C. In combination with vacuum like a thermus it has a heat conductivity of only 0.006 W/mK. This value is increased only by a factor of 3 without vacuum. Beyond its primary function of thermal enclosure it is a protective container for all fault or accident conditions.
4. Barrier by the battery controller. The battery controller supervises the battery and stops operation in any undesired situation.

10.2.7 Recycling

Nowadays any product that is introduced to the market has to be recycled at the end of its usage. ZEBRA batteries are dismantled. The box material is stainless steel and SiO_2 , both of which are recycled by established processes. The cells contain Ni, Fe, salt, and ceramic. For recycling they are simply added to the steel melting process of the stainless steel production. Nickel and iron are contributed to the material production and the ceramic and salt is welcome to form the slag. The recycling is certificated and cost effective.

10.2.8 Applications

The ZEBRA battery system is designed for electric vehicles (Figure 10.12) which require a balance of power to energy of about 2, e.g., a 25 kWh battery has about 50 kW peak power. Other applications are electric vans, buses, and hybrid buses with ZEV range (Figures 10.13 and 10.14).

The present generation of ZEBRA batteries is not applicable for hybrid vehicles that have a small battery of about 3 kWh but high power up to 60 kW (a power to energy ratio of 15 to 20). Recently also prototypes for stationary applications have been constructed. These have great advantages in hot climates and for frequent cycling, where the lifespan of conventional batteries is reduced such that the two- to three-times higher price of ZEBRA batteries is overcompensated by its much longer life, resulting in lower life cycle cost and avoiding the exchange of batteries. For uninterrupted power source (UPS) applications the float voltage of 2.61 V/cell for ZEBRA batteries has been established.

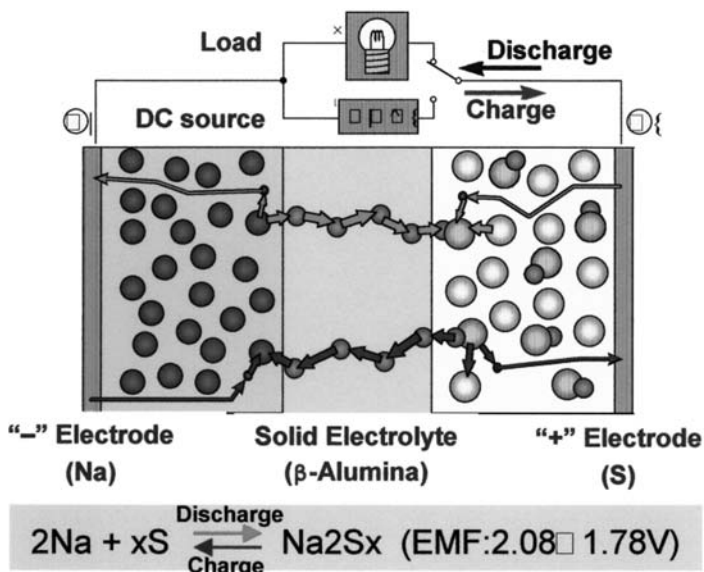
10.3 NaS BATTERY

10.3.1 Technology

Sodium-sulfate batteries use metallic sodium and sulfur, both in the liquid state, for the electrode material and are assembled in the charged state. The electrolyte and separator is β'' -Al₂O₃-ceramic as in the ZEBRA battery (Fig. 10.13). During discharge the sodium is conducted through the β -alumina to react with sulfur to form Na₂S_n with $3 < n < 5$. For charge the same reaction is reversed. The main components of the NaS cell are shown in Figure 10.14. The negative pole is connected to the sodium container in the center of the cell, which is made out of stainless steel. This container has a small hole at the bottom which is designed to limit the sodium flow in case of overheating or overvoltage. The container is



Figure 10.12 Electric vehicles.



- NA, elemental sodium
- NA^+ , sodium ion
- S, elemental sulfur
- Na_2S_x , sodium polysulfide
- e^- , electron

Figure 10.13 NaS battery chemistry.

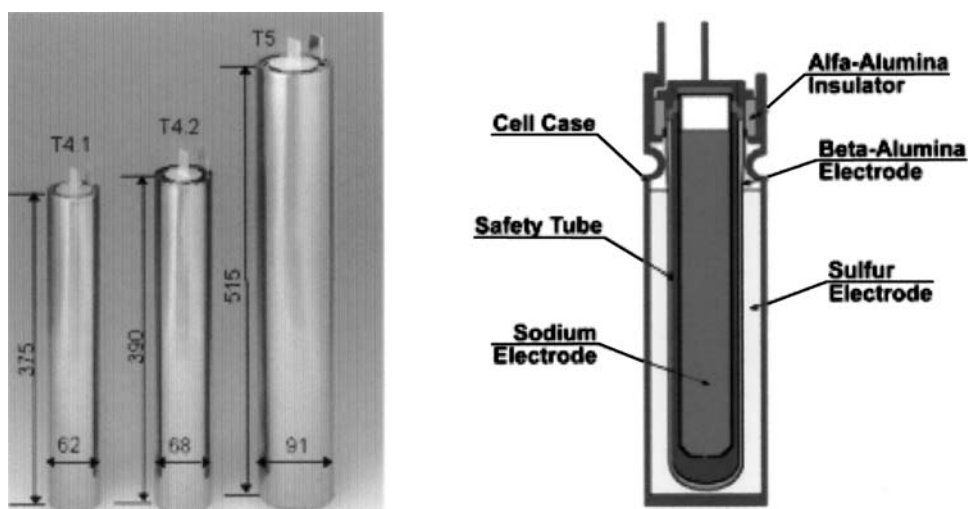


Figure 10.14 NaS cell components. (From Ref. 10.)

surrounded by a deep drawn safety insert in aluminum, which has a tight fit to the inner shape of the β - Al_2O_3 tube in order to form a capillary gap for the wetting of the ceramic electrolyte. The ceramic tube is hermetically sealed with a thermocompression-bound (TCB) seal to the negative and positive poles, which are formed by the cell case. The sulfur is contained in the space between the ceramic tube and the cell case. Sulfur and, in a partially discharged state, polysodium sulfur are both electrically nonconductive so that a carbon-felt is integrated for conductivity. The cell case is made out of aluminum and the inside coated with a conductive layer for corrosion protection.

The NaS cells now being produced by the Japanese company NGK Insulators have 632 Ah and 340 kWh/m³. 320 of such cells have been connected in parallel and in series to form modules with 375 kWh and 3500 kg weight. These modules are connected together for load leveling and power quality plants with up to 6 MW/48 MWh. About 50 of such plants are in operation, the oldest since 1992.

Initially NaS batteries were developed for mobile and stationary applications. But during abuse testing and simulations of heavy accidents the sodium and sulfur reacted in an uncontrolled way and toxic gas was identified. For this reason NaS batteries are no longer considered for mobile applications, but only for stationary load leveling where damage due to accidents need not be considered (Figure 10.15).

10.4 LITHIUM-ION BATTERY

10.4.1 Technology

Lithium is a light and very reactive metal so that it is attractive for electrochemical energy storage if a stable electrolyte can be found. There are different Li salts used that are solved in nonaqueous solvents like methyl acetate or methyl formate. The positive electrode is LiCoO_2 , which is mostly used today, but for car battery applications it is too expensive. LiNiO_2 and LiMn_2O_4 are under investigation and should lead to lower cost, but have less energy density.

The negative electrode is intercalated carbon that can store Li up to C_6Li . The principle is shown in Figure 10.16.

Rechargeable Li-ion batteries were introduced to the market for consumer products like mobile phones and notebooks. For electric vehicles up to now only experimental cars have been demonstrated [12]. The main open tasks are related to safety under abusive conditions and cost. Li-ion batteries can be designed for high power or high energy (Table 10.1). It can be expected that they will be a candidate for 42-V car systems as soon as safety and cost levels are satisfactory.

10.5 LITHIUM-POLYMER BATTERY

Lithium-polymer batteries (LPBs) use intercalated carbon for the negative electrode, a polymer electrolyte, and metallic lithium that is deposited as a thin film as the negative electrode. This battery type is under development but not yet introduced to the market for electric vehicles.

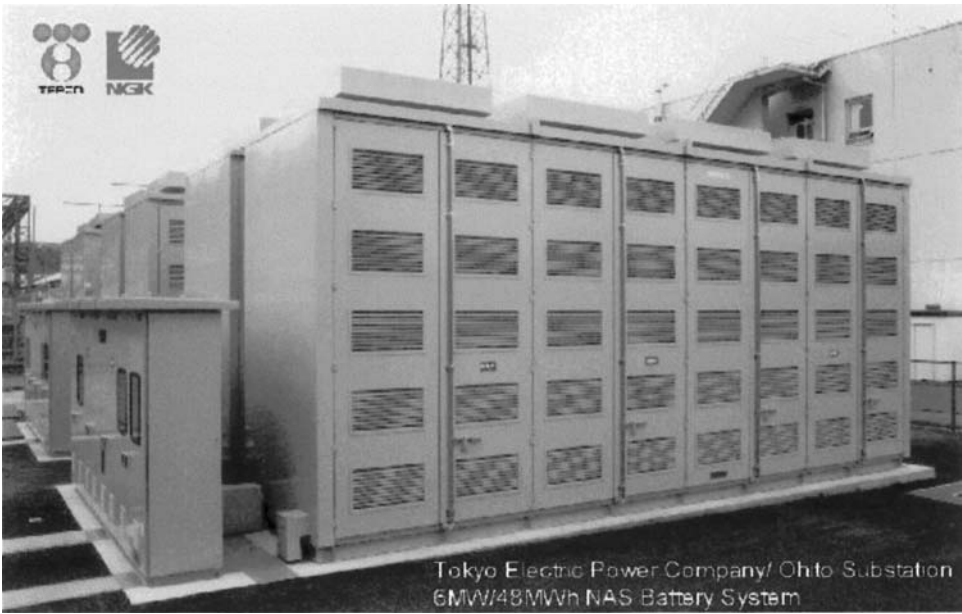


Figure 10.15 NaS battery load levelling plant built by NGK Insulators (Japan).

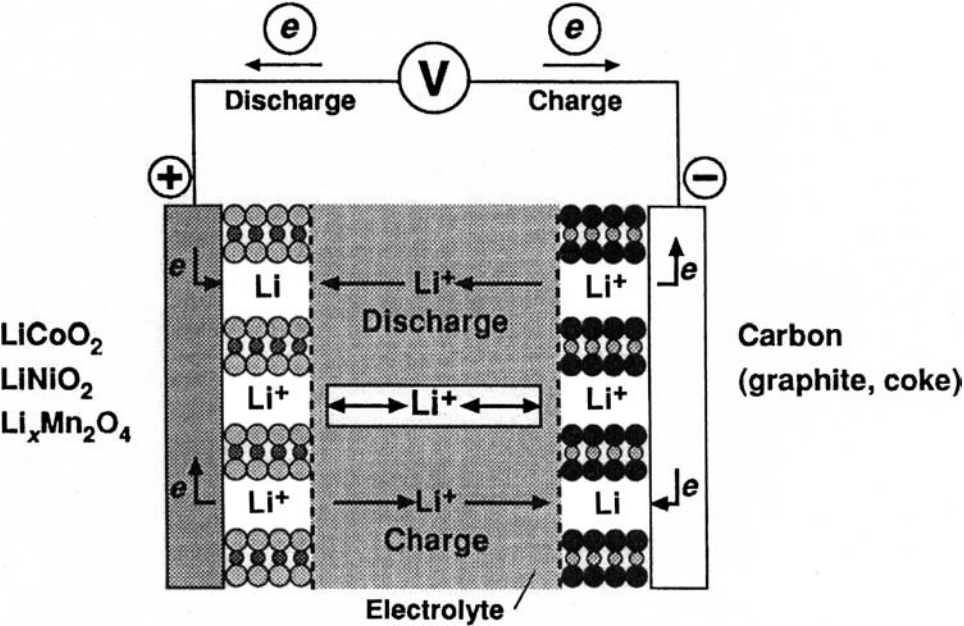


Figure 10.16 Schematic of a Li-ion battery. (From Ref. 8.)

10.6 OTHER BATTERY SYSTEMS

For completeness Zn-halogen and redox batteries should be mentioned. These types of batteries have the advantage of the ability to separate the electrolyte from the storage of an electrode material that is liquid and can be stored in tanks (Figure 10.17). By this means power and energy content are independent from one another. For mobile applications this battery type no longer has any relevance due to safety concerns. In case of accident the liquid could be spilled out and, e.g., bromine would be liberated. But for stationary applications the possibility to store large quantities of energy in tanks separate from the power-determining electrolyte justifies the leak detection effort. Therefore, redox battery systems are still under consideration for stationary electric energy storage.

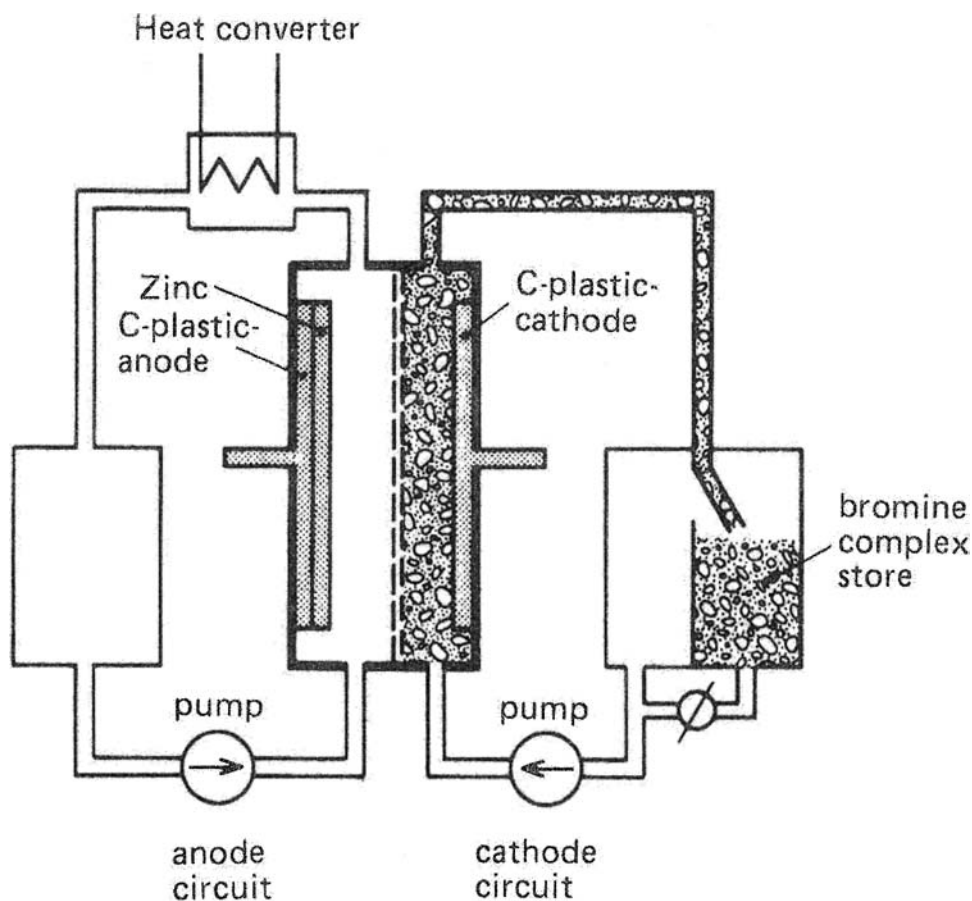


Figure 10.17 Principle of the Zn/halogen accumulator with two different electrolyte circuits.

10.7 BATTERY OVERVIEW

10.7.1 Minimum Requirements for EV Batteries

From mobility studies [13] it has been well established that the average distance driven in cars per day is 40–60 km and 40–60 miles in the United States, subdivided into 3.8 trips. At least during the introduction phase it cannot be assumed that public charging stations will be available and that the drivers are willing and able to plan their trips with such accuracy and detail that charging between trips during the day is acceptable. Therefore, the strategy for the introduction of electric vehicles can only be to charge at night and drive during the day. Therefore, the autonomy of EVs has to be 60 km (60 miles in the United States) plus a reserve of 30 to 40%. EV fleet tests [14] have shown that normal EV drivers use 60 to 70% of their capacity independent of the range because they feel comfortable only with a sufficient reserve. These above facts lead to the requirement of at least a 100-km range under any conditions, like severe weather. The other fundamental requirement is a top speed of 100 km/h because vehicles for urban traffic also use motorways where trucks are driving at 80 km/h and it must be possible to overtake them with a sufficient speed difference.

Figure 10.18 shows the range of a typical EV depending on its speed for three typical specific energy values of batteries. From this it is obvious that the battery of a marketable electric vehicle has to have about 100 Wh/kg as its minimum.

For the other requirements of safety, vibration resistance, climate, etc., the reader is referred to Chapter 4, Sec. 4.4.

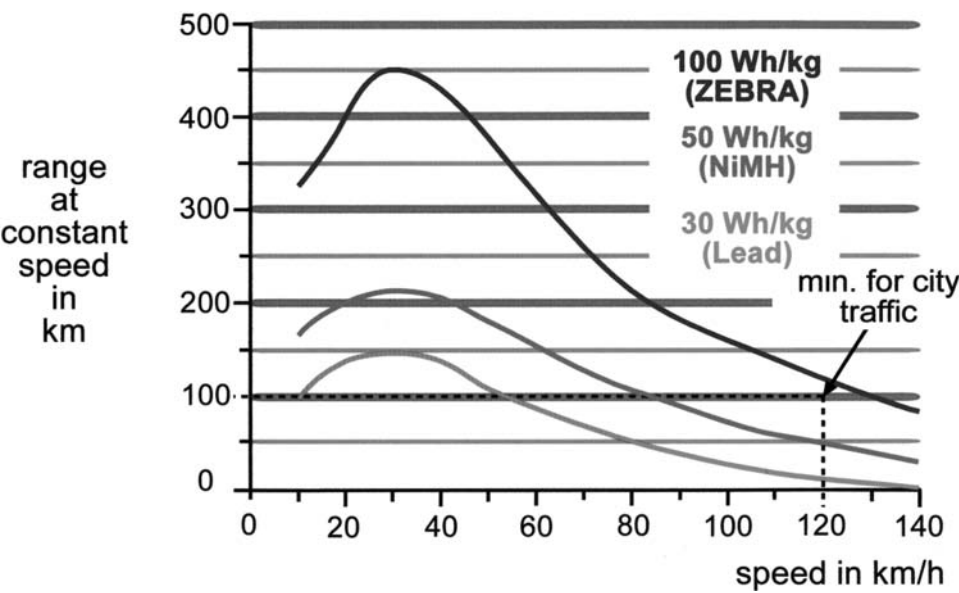


Figure 10.18 Range of electric vehicles depending on the speed and battery specific energy.

10.7.2 ZEV Life Cycle Costs Start to Be Competitive

A car for urban use drives typically 10,000 to 15,000 km per year for about 10 years so that the operating cost for 150,000 km should be considered. All investigations on vehicle cost indicate that the electric car cost without the battery is equal to or even a little less than a comparable thermal engine car, whereas for operation electricity is cheaper than fuel. Figure 10.19 shows that EVs reach breakeven cost at a fuel price of 1 EUR/L and a battery price of 300 Euro/kWh. For ZEBRA batteries the necessary battery life of 10 or more years and 1000 or more nameplate cycles have been demonstrated. It can be expected that the battery will last as long as the vehicle. Another option is battery rental, for which the monthly rental fee is paid out of the energy cost difference between electricity and fuel. For electric vehicles the maintenance costs (no oil replacement, nearly no brake disk exchange due to electric regenerative braking, no exhaust pipe replacement) irrespective of the energy costs and the assurance costs are less than for conventional cars.

These conditions are beginning to be realistic due to rise of crude oil price and the beginning of series production of at least the ZEBRA battery. Now electric vehicles are starting to become an option for urban traffic, about 100 years after their first period of success.

10.8 FUEL CELLS

Fuel cells have been known about for more than 150 years [15]. W. R. Grove operated the first fuel cell in his laboratory by 1842, 17 years before G. Planté built the first lead-acid accumulator. However, they never could compete with the thermomechanical process for electricity production. But they are being rediscovered nowadays because they offer the possibility to convert chemical energy to electricity

Fuel consumption of thermal engine car 5l/100km

Electricity consumption of ZEV 20 kWh/100 km

Electricity price for night charge 0.07 EUR/kWh

Battery charger efficiency 90%

Battery nameplate cycle life 1500

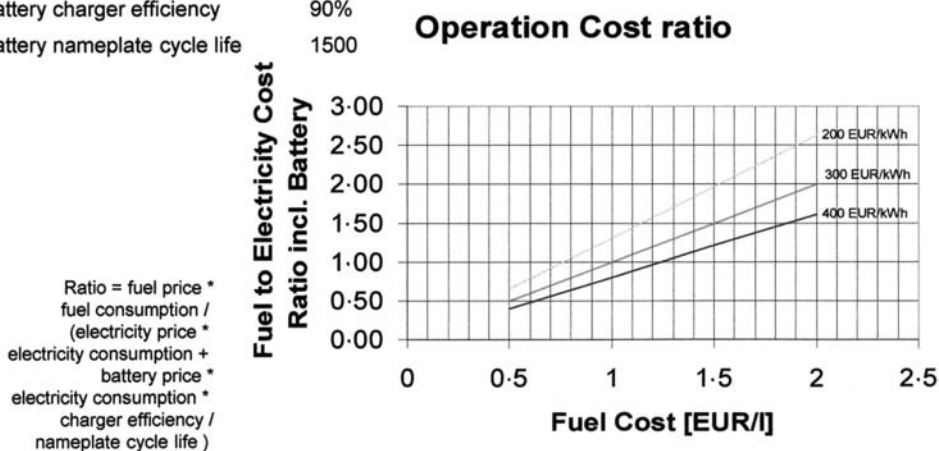


Figure 10.19 EV battery and electricity cost compared to fuel cost.

Table 2 Different fuel cell types.

Type	Electrolyte	Ionic conduction	Fuels	T (°C)
SOFC	ZrO ₂ ceramic	O ²⁻	CH ₄ , CO, H ₂	500–900
MCFC	NaCO ₃ liquid	CO ³⁻	CO, H ₂	650
AFC	KOH liquid	(OH) ⁻	Pure H ₂	70
PAFC	H ₂ PO ₄ liquid	H ⁺	H ₂	200
PEM	Polymer solid	H ⁺	Pure H ₂	80

directly unconstrained by the Carnot efficiency. The different types of fuel cells are listed in Table 10.2.

The main effort of recent development work and investment is concentrated on the industrialization of PEM and SOFC type fuel cells. Both have advantages and disadvantages: PEM fuel cells are operated at a convenient low temperature of 80 °C, can be started from ambient temperature, and the technology is mature for first field tests. Its important disadvantage is that it operates only with pure H₂ as its fuel, which is not a generally available secondary energy with an existing infrastructure. The use of PEM fuel cells for electric vehicles requires the solution of another issue, the production, distribution and storage of hydrogen in large scale. SOFCs operate with any fuel because its ceramic electrolyte conducts oxygen ions that “burn” any fuel. The disadvantage is the high operating temperature of 500 to 900 °C. A small unit for about 5 kW is under development.

Actually there is a large interest in fuel cells and many updated publications are available [16,17]. For this reason a more detailed presentation of this subject is not attempted in this chapter.

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