Functional principle and the main components of lithium and Li-ion batteries (primary-, secondary)

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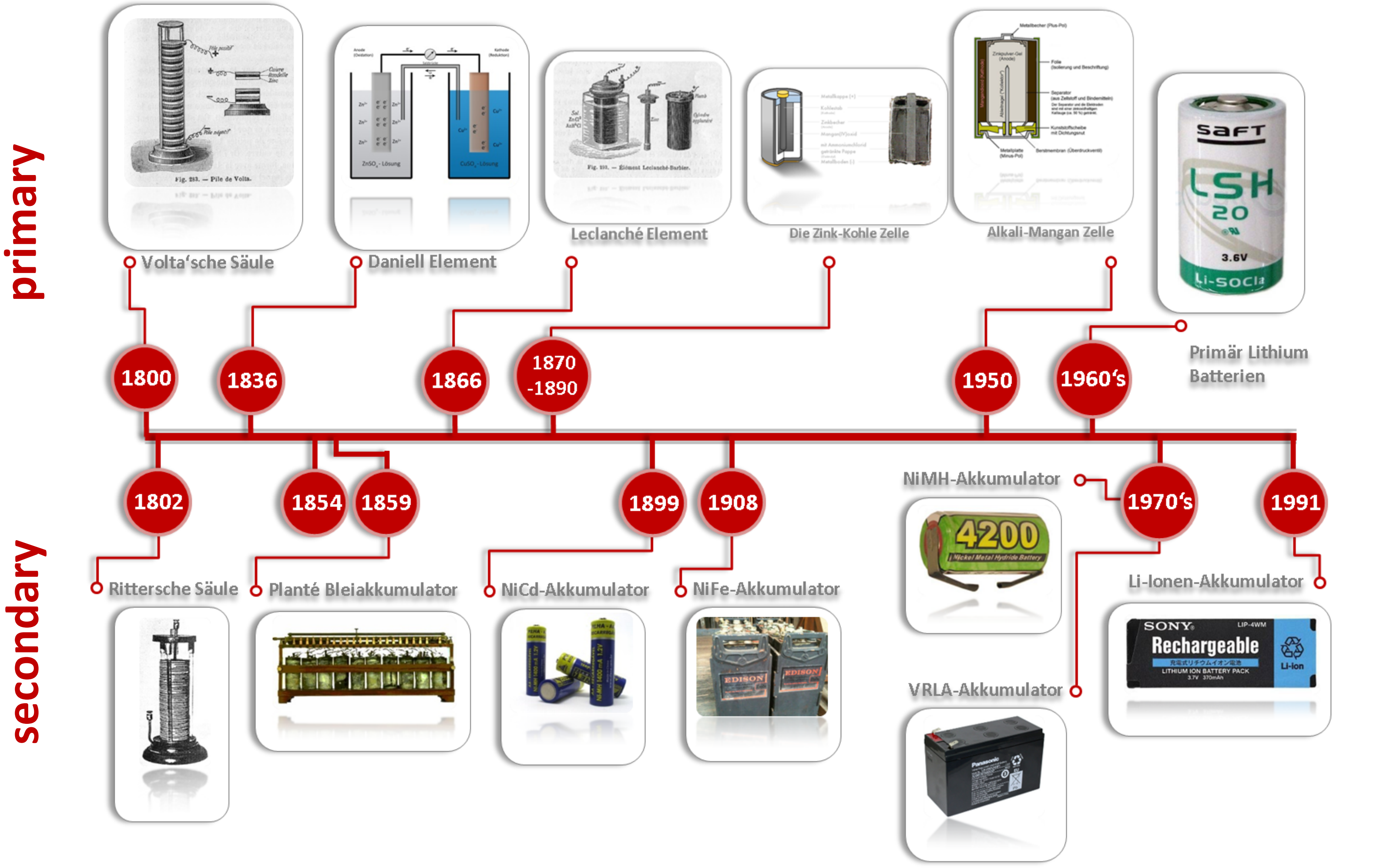
Short history of the galvanic cells



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Short history of the galvanic cells

Year	Inventor	Activity
1600	William Gilbert (UK)	Establishment of electrochemistry study
1745	Ewald George von Kleist (Netherlands)	Invention of Leyden jar. Stores static electricity
1791	Luigi Galvani (Italy)	Discovery of “animal electricity”
1800	Alessandro Volta (Italy)	Invention of the voltaic cell (zinc, copper disks)
1802	William Cruickshank (UK)	First electric battery capable of mass production
1820	André-Marie Ampère (France)	Electricity through magnetism
1833	Michael Faraday (UK)	Announcement of Faraday’s law
1836	John F. Daniell (UK)	Invention of the Daniell cell
1839	William Robert Grove (UK)	Invention of the fuel cell (H_2/O_2)
1859	Gaston Planté (France)	Invention of the lead acid battery
1868	Georges Leclanché (France)	Invention of the Leclanché cell (carbon-zinc)
1899	Waldmar Jungner (Sweden)	Invention of the nickel-cadmium battery
1901	Thomas A. Edison (USA)	Invention of the nickel-iron battery
1932	Schlecht & Ackermann (D)	Invention of the sintered pole plate
1947	Georg Neumann (Germany)	Successfully sealing the nickel-cadmium battery
1949	Lew Urry, Eveready Battery	Invention of the alkaline-manganese battery
1970s	group effort	Development of valve-regulated lead acid battery
1990	group effort	Commercialization of nickel-metal-hydride battery
1991	Sony (Japan)	Commercialization of lithium-ion battery
1994	Bellcore (USA)	Commercialization of lithium-ion polymer
1996	Moli Energy (Canada)	Introduction of Li-ion with manganese cathode
1996	University of Texas (USA)	Identification of Li-phosphate ($LiFePO_4$)
2002	University of Montreal, Quebec Hydro, MIT, others	Improvement of Li-phosphate, nanotechnology, commercialization

Short history of the Li-ion batteries

1970's: commercialisation of the first non-rechargeable lithium battery

1980's: development starts on rechargeable Li-ion cells (with metallic Li)

1991: commercialisation of rechargeable Li-ion cells (Sony)

1994: commercialisation of rechargeable Li-ion polymer cells, „LiPo" (Bellcore)

1996: Introduction of the lithium-manganese-oxide (LMO) cathode (Moli Energy)

1996: Introduction of the lithium-iron-phosphate (LFP) cathode material (Univ. Texas)

Advantages of the Li-ion technology

- maintenance-free
- no „memory effect“
- no self-discharge
- >3,6V cell voltage → highest energy density
- available as „energy cell” and „power cell”
- fast charge/discharge is possible
- high efficiency (i.e., Coulombic efficiency)
- broad temperature range (-20°C - +60°C)
- flat voltage profile

Disadvantages of the Li-ion technology

- active charge- and discharge safety procedures, „cell balancing” needed
- Manufacturing complex, highly cost-intensive, expensive/high price
- possible cell opening on cell abuse (mechanic-, electric-, thermal abuse)
- strict transportation provisions

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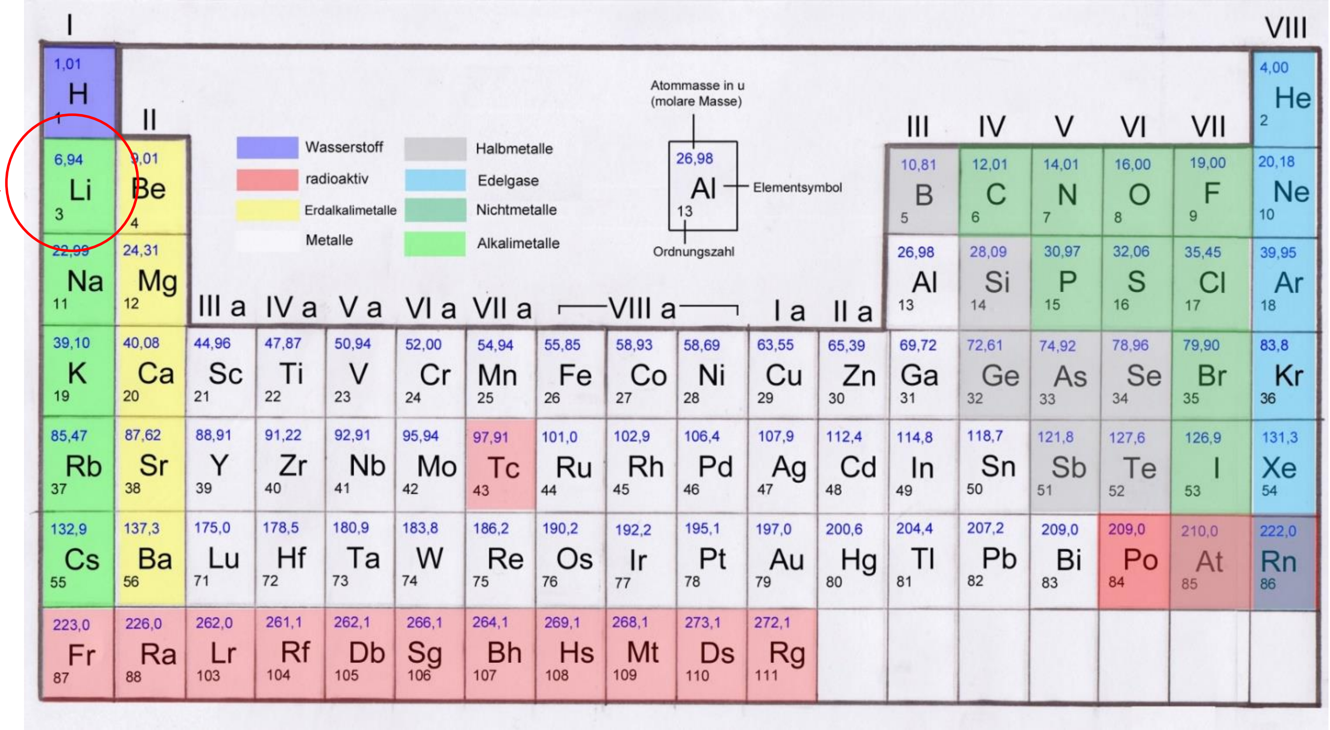
Why lithium?

Alkali metal
Atomic number: 3
Atomic weight: 6,94
Spec. gravity: 0,53 g/cm^3



Name origin:
Ancient Greek
λίθος (*líthos*) =
„Stein“

- lightest metal on Earth (lightest from all solid element)
- highest electrochemical potential
- highest specific energy is achievable

However very reactive!!!

Reaction between lithium and water

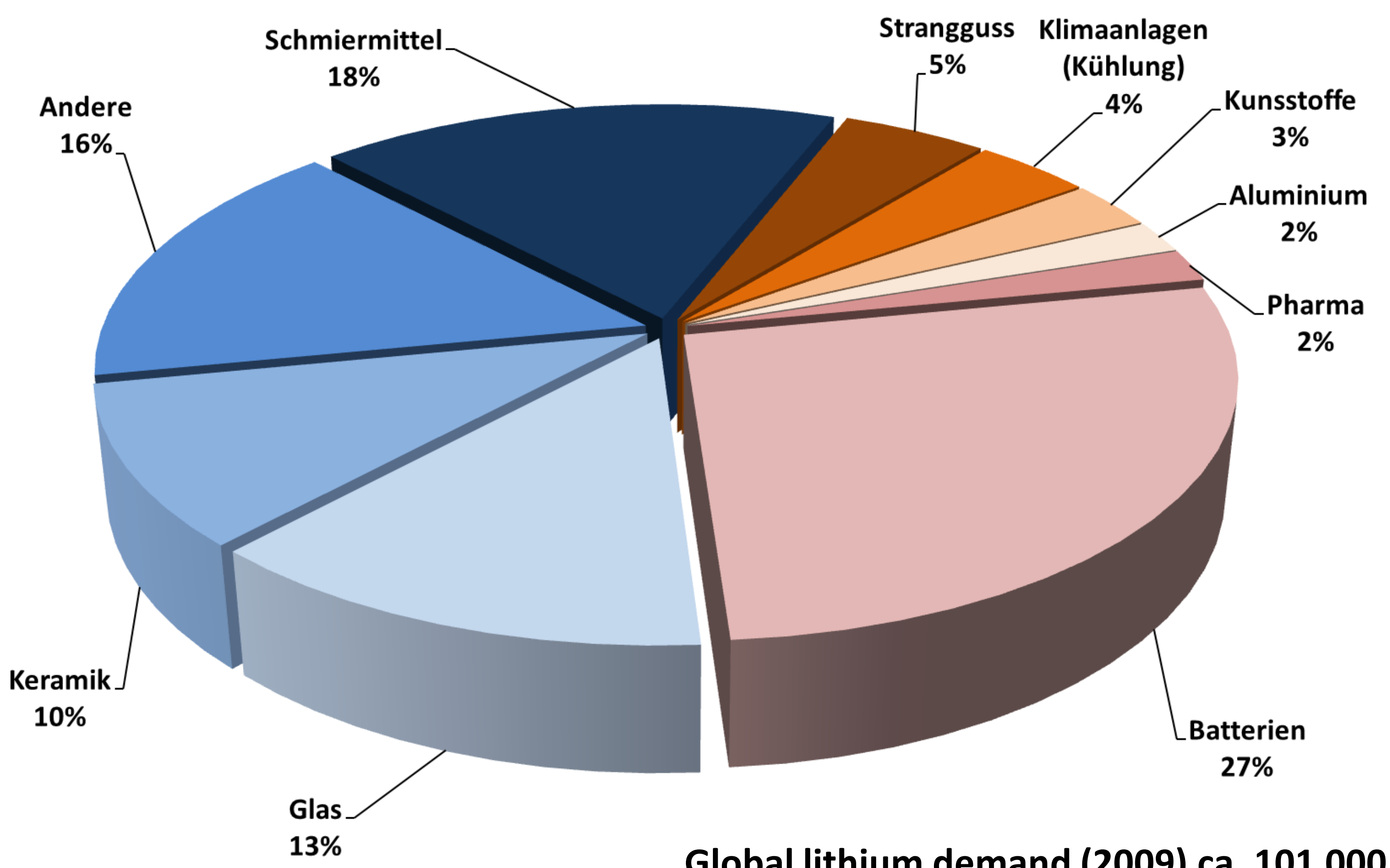
Quelle: http://www.dlt.ncssm.edu

Working in glovebox

http://www.ifam.fraunhofer.de/en/Bremen/Shaping_Functional_Materials/Equipment.html

Working in dry room: RH% <0,3

Range of application of lithium



Global lithium demand (2009) ca. 101 000 t

(for batteries ca. 27 000 tons)

Nissan Leaf (Full EV)

ca. 4 kg „lithium“ in the battery pack

http://cleantechnica.com/files/2014/07/leaf-battery1.jpg

„The lithium triangle“

Chile, Bolivia, Argentina

Economist.com

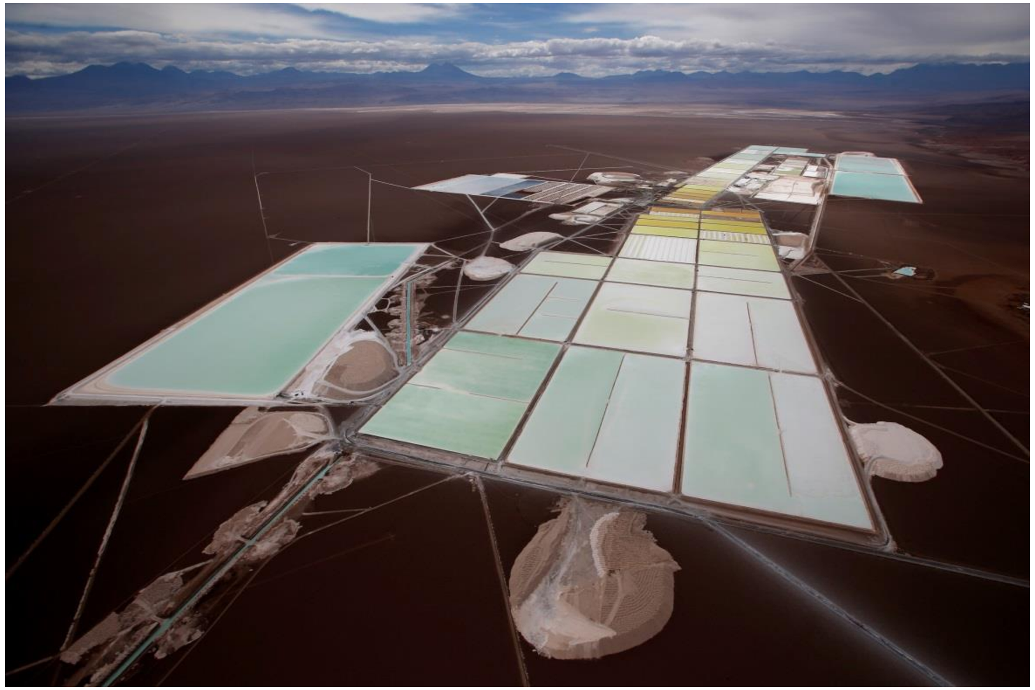
Production of lithium

- 70% from salt water (brine) (residual from rocks)
- 750 t brine → ~1 t Li (in 24 month procedure)

„The lithium triangle“ - Chile, Bolivia, Argentina

2nd largest salt flat on Earth and
World-wide largest lithium deposit, i.e., about 25% of the Earth’s resources (!)

Rockwood Lithium Plant, Antofagasta , Atacama Wüste, Chile



Rockwood Lithium, Antofagasta, Atacama Wüste, Chile

Rockwood Lithium, Antofagasta, Atacama Wüste, Chile

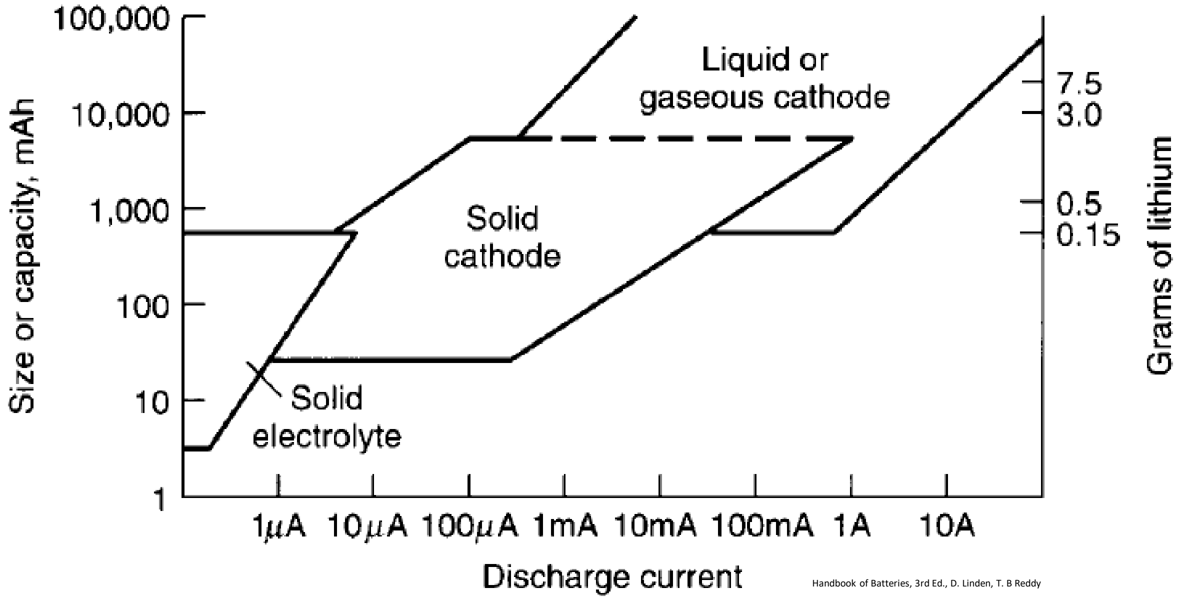
Bildnachweis: http://blogs.reuters.com/photographers-blog/2013/04/05/the-lithium-triangle/

Primary lithium batteries

Classification of the primary lithium systems

Primary lithium batteries

- **liquid cathode**
- **solid state cathode**
- **solid state electrolyte**



Handbook of Batteries, 3rd Ed., D. Linden, T. B Reddy

Primary lithium batteries with liquid cathode

Lithium-Schwefeldioxid-Zelle, Li/SO_2

Anode: Lithium Metall

Kathode: SO_2 / hochporöser Kohlenstoff

Elektrolyt: SO_2/Acetonitril/LiBr

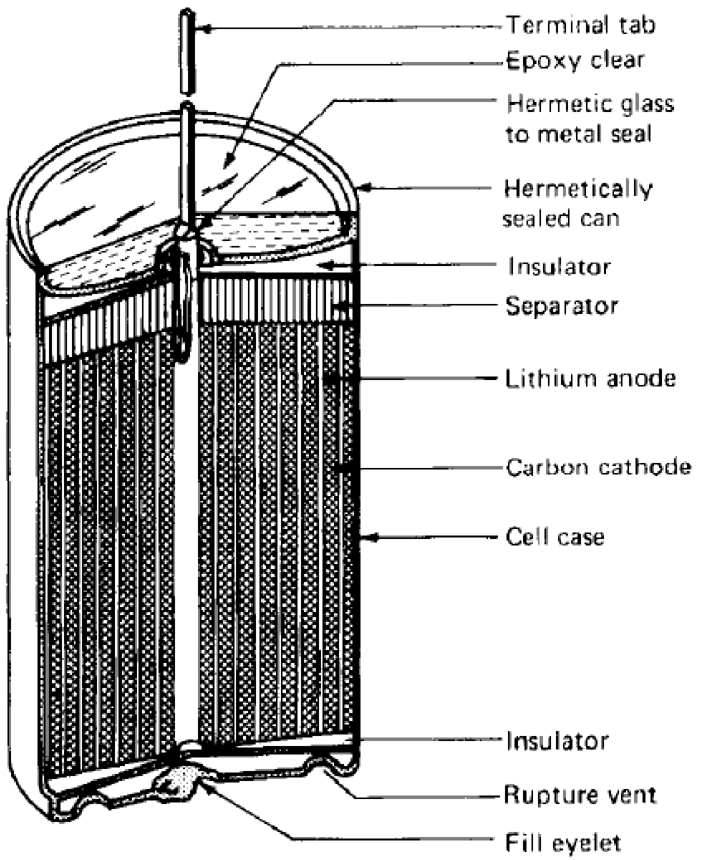
Ruhespannung: 3,0 V

ca. 260 Wh/kg, 415 Wh/l

Hochstrom/Niedrig-temperatur Anwendungen

Zelle unter Druck: 3-4 Bar

Temperaturbereich: -40 - +55°C



Reaktion (Gesamt):

$$\overset{0}{2\,Li} + 2\,\overset{+4}{S}O_2 \rightarrow \overset{+1}{Li_2}\overset{+3}{S_2}O_4 \text{ (Lithium Dithionit)}$$

Meistens „kathodenlimitiertes“ Entladeprozess

Primary lithium batteries with liquid cathode

Lithium-Thionylchlorid-Zelle, Li/$SOCl_2$

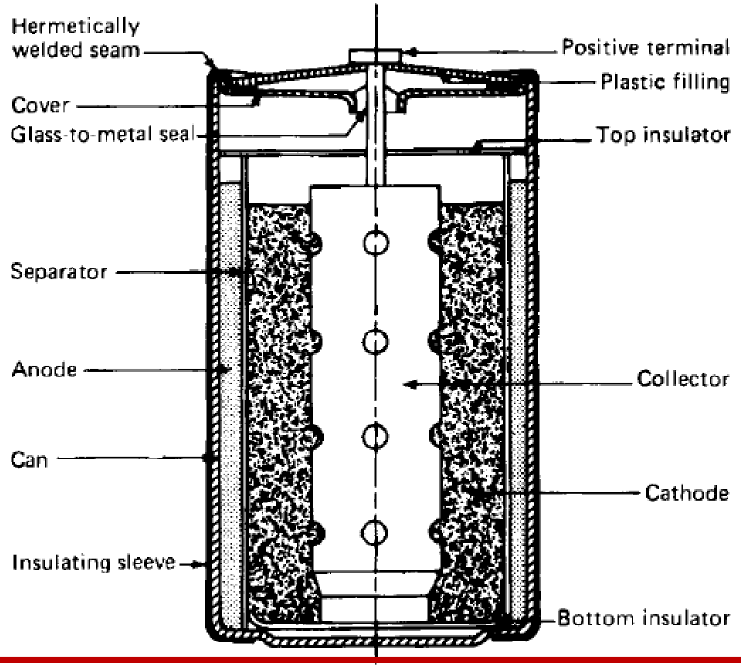
Anode: Lithium Metall

Kathode: $SOCl_2$ / hochporöser Kohlenstoff

Elektrolyt: $SOCl_2$/$LiAlCl_4$ ($LiGaCl_4$)

Ruhespannung: 3,6 V

Baugrößen: 400 mAh → 10 000 Ah (!)



www.varta-microbattery.com

Reaktion (Gesamt):

$$\overset{0}{4\,Li} + 2\,\overset{+4}{S}OCl_2 \rightarrow 4\,\overset{+1}{Li}Cl + \overset{0}{S} + SO_2$$

Lithium-Sulfurylchlorid-Zelle, Li/SO_2Cl_2

Anode: Lithium Metall

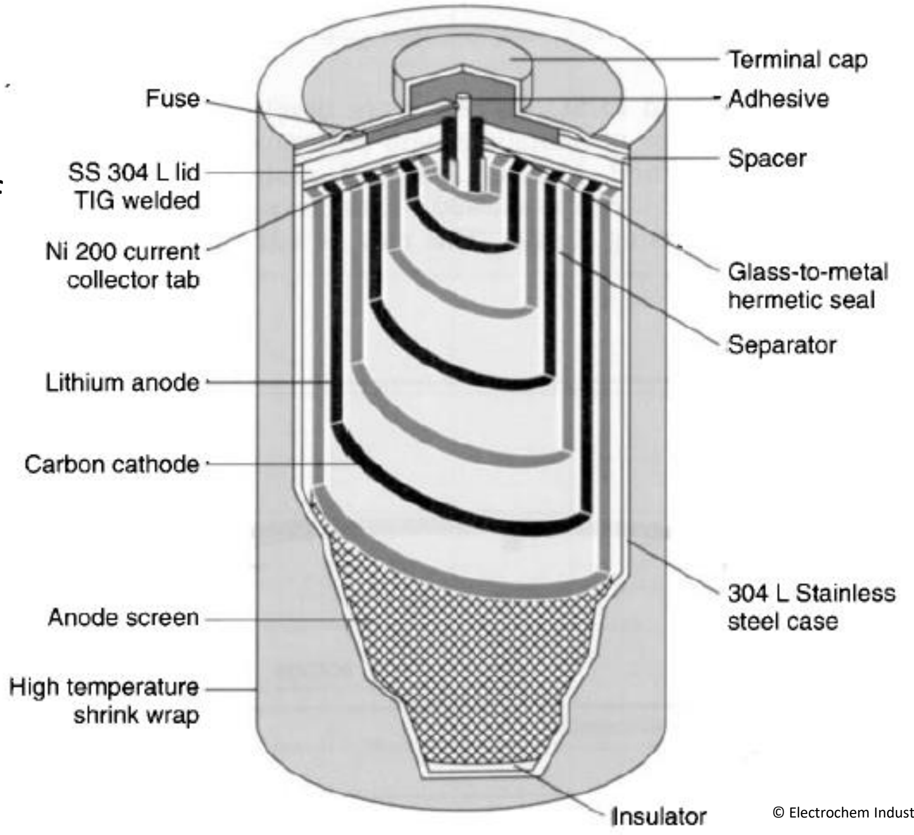
Kathode: SO_2Cl_2 / hochporöser Kohlenstoff

Elektrolyt: $SOCl_2/LiAlCl_4$

Ruhespannung: 3,90 V (3,95V)

Temperaturbereich: -30 - +90°C

Additive: Cl_2 (für höhere U (3,95V), Wh/kg, Wh/l, sicherer Betrieb)



© Electrochem Industries

Reaktionen

Anode: $Li \rightarrow Li^+ + e^-$

Kathode: $SO_2Cl_2 + 2e^- \rightarrow 2\,Cl^- + SO_2$

Summe: $2\,\overset{0}{Li} + \overset{+6}{S}O_2Cl_2 \rightarrow 2\,\overset{+1}{Li}Cl + \overset{+4}{S}O_2$

Lithium-Eisensulfide-Zelle, Li/FeS_2

Anode: Lithium Metall

Kathode: FeS_2

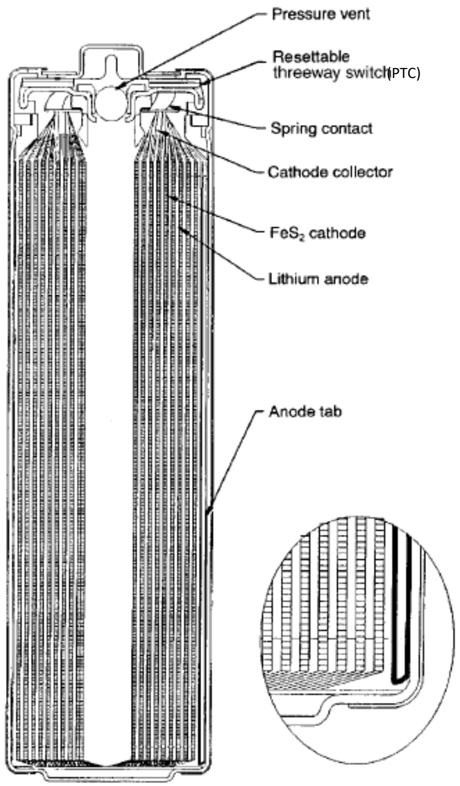
Elektrolyt: LiI/Solvent

Ruhespannung: 1,80 V

Nennspannung: 1,50 V

Temperaturbereich: -40 - +60°C


Pyrit



Reaktionen

$$\text{Anode: } 4\,Li \rightarrow 4\,Li^+ + 4e^-$$

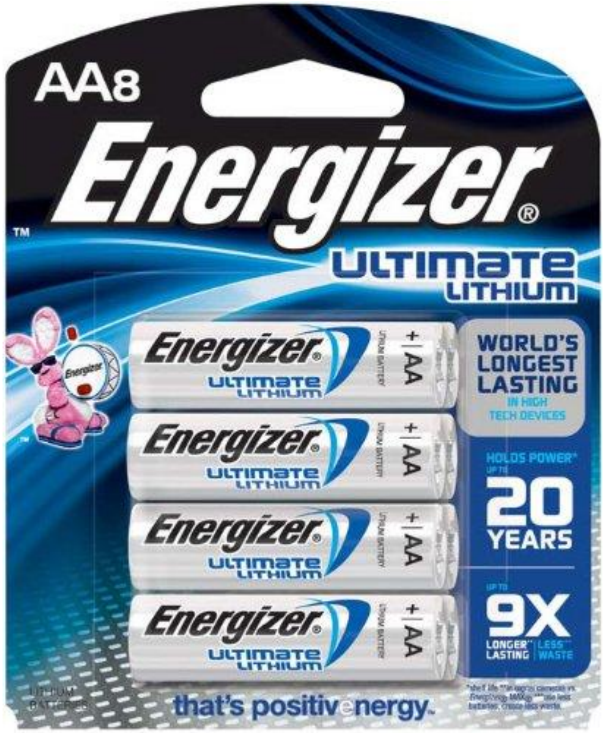
$$\text{Kathode: } FeS_2 + 4e^- \rightarrow Fe + 2\,S^{2-}$$

$$\text{Summe: } 4\,\overset{0}{Li} + \overset{+2}{Fe}S_2 \rightarrow \overset{0}{Fe} + 2\,\overset{+1}{Li}_2S$$

Gute Hochstrom/Niedrig-temperatur Leistungsfähigkeit

Primary lithium batteries with solid state cathode

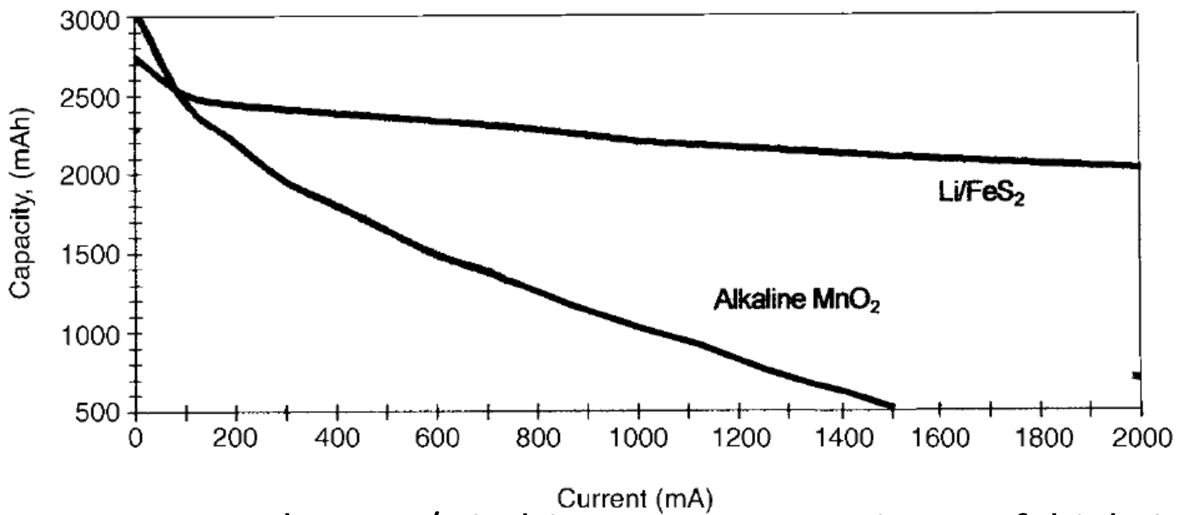
Lithium-Eisensulfide-Zelle, Li/FeS_2



conrad.de

Entladestrom vs. Entladekapazität, AA-Größe, @ 21°C

(Eveready Battery Co, Inc.)



Bessere Hochstrom/Niedrig-temperatur Leistungsfähigkeit als Zn/MnO_2 Zellen

Primary lithium batteries with solid state cathode

Lithium-Manganoxid-Zelle, Li/MnO_2

Anode: Lithium Metall

Kathode: MnO_2

Elektrolyt: $LiClO_4$ in PC/1,2-Dimethoxyethane

Nennspannung: 3,0 V (cut-off: 2 V)

Ruhespannung: 3,3 V

Temperaturbereich: -20 - +55°C

Spezifische Energie: 230 Wh/kg

Energiedichte: 530 Wh/l

+ Kein „voltage delay“
+ Gute Lagerfähigkeit
  (Selbstentladung <1%/Jahr)

Reaktionen

Anode: $x\,Li \rightarrow x\,Li^+ + x\,e^-$

Kathode: $MnO_2 + x\,Li^+ + x\,e^- \rightarrow Li_xMnO_2$

Summe: $x\,\overset{0}{Li} + \overset{+4}{Mn}O_2 \rightarrow \overset{+1}{Li_x}\overset{+3}{Mn}O_2$

(Interkalation)

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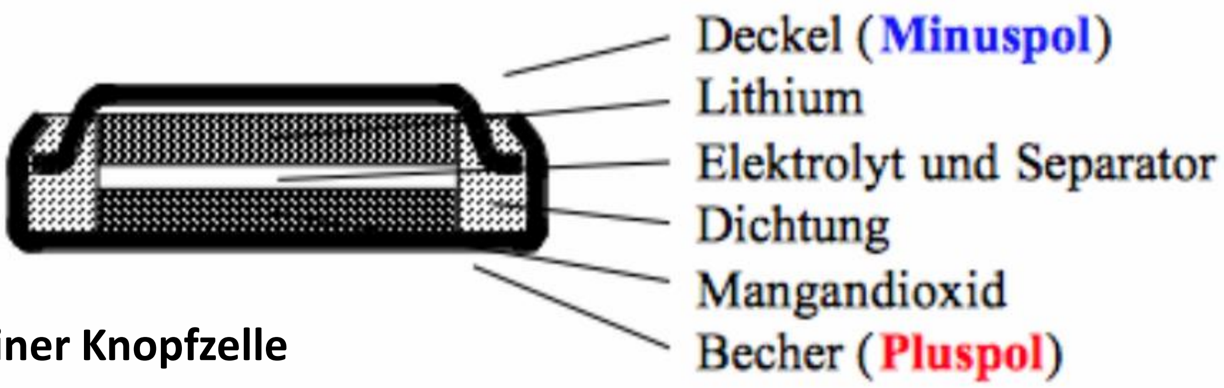
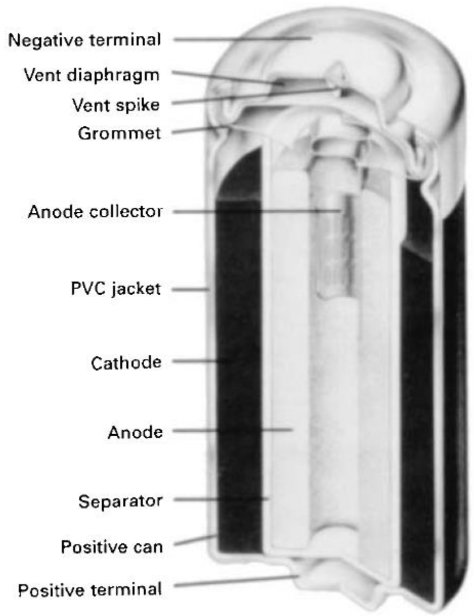
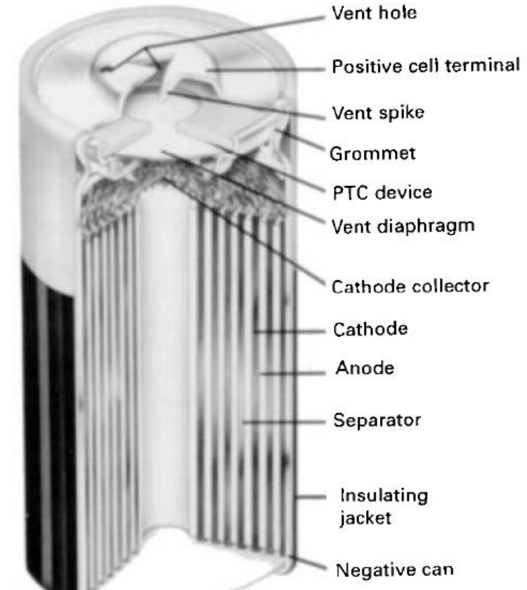
Lithium-Manganoxid-Zelle, Li/MnO_2



Prinzipieller Aufbau einer Knopfzelle

Bauformen:
- Knopfzelle
- Massezelle
- Wickelzelle

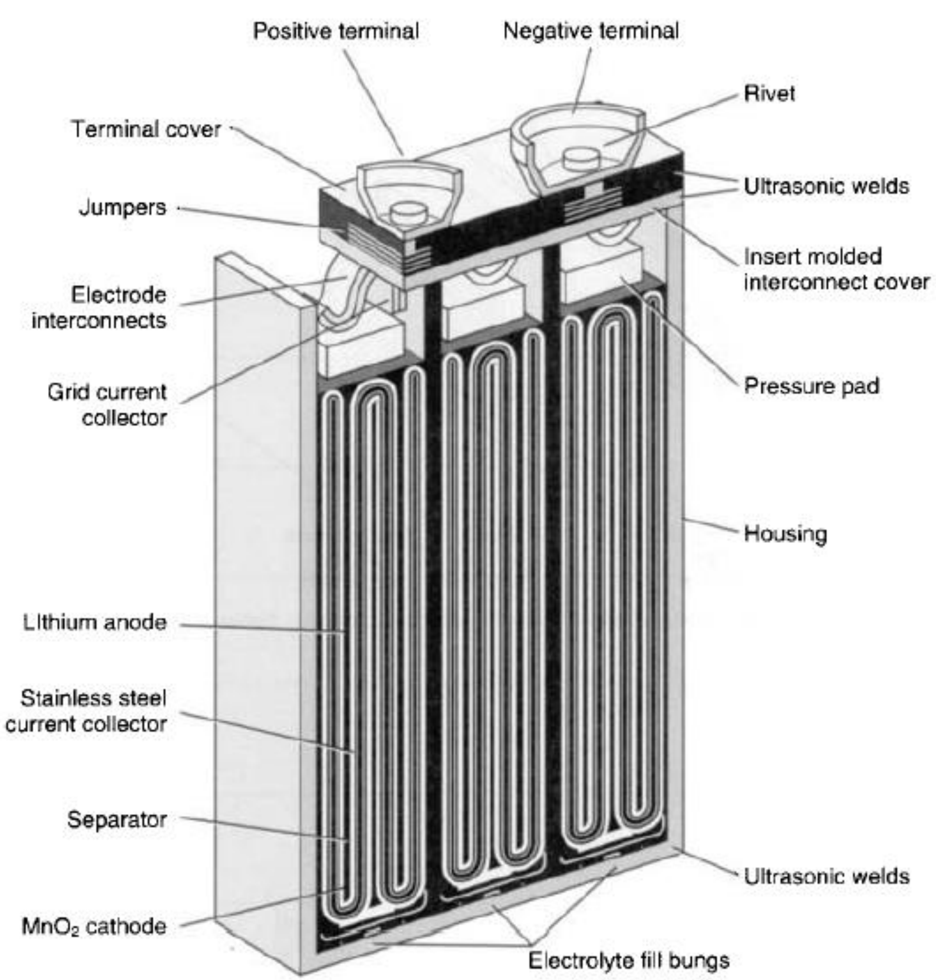




Handbook of Batteries, 3rd Ed., D. Linden, T. B Reddy

Lithium-Manganoxid-Zelle, Li/MnO_2

9V-Multizelle-Batterie



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Lithium-Kohlenstoff-Monofluorid-Zelle, $Li/(CF)_x$

Anode: Lithium Metall

Kathode: Poly-Kohlenstoff Monofluorid $(CF)_x$

Elektrolyt: $LiBF_4$ /$LiClO_4$ in PC/Dimethoxyethane

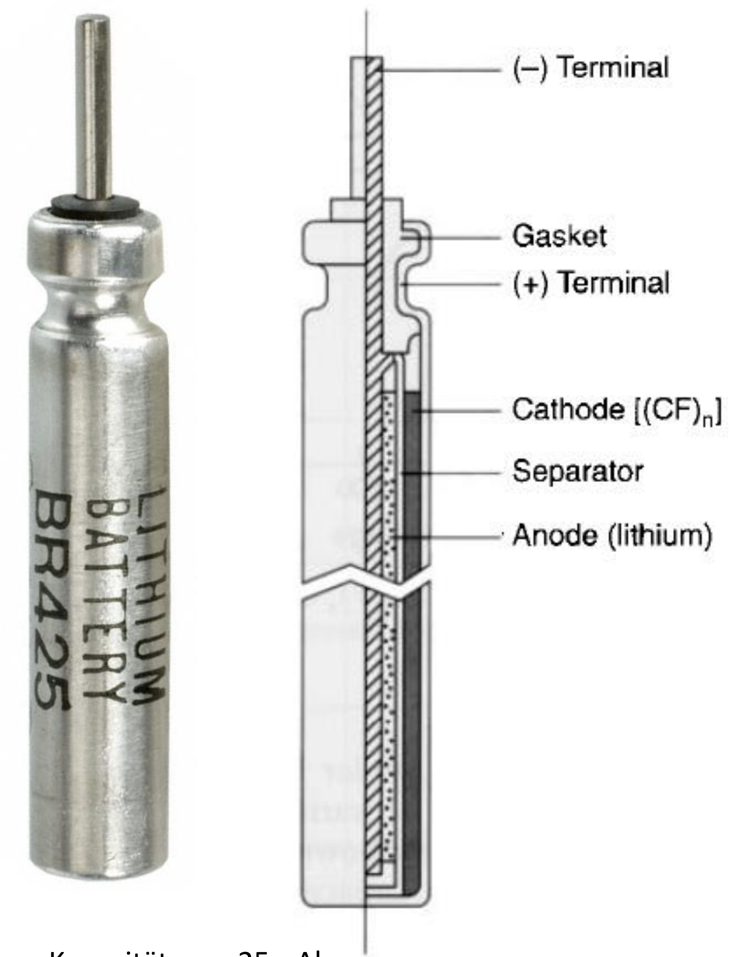
Nennspannung: 2,5 - 2,7 V

Ruhespannung: 3,2 V

Lagerfähigkeit: 10+ Jahre

Spezifische Energie: 250 Wh/kg (590 Wh/kg, Großformat)

Energiedichte: 635 Wh/l (1050 Wh/l, Großformat)



Kapazität	25mAh
Größe	DxH: 4,2 x25,9 mm
Abschlussart	Leiterplattenstift
Entladerate	500µA
Gewicht	0,57g

Reaktionen

Anode: $x\,Li \rightarrow x\,Li^+ + x\,e^-$

Kathode: $(CF)_x + x\,e^- \rightarrow x\,C + x\,F^-$

Summe: $x\,\overset{0}{Li} + (\overset{-1}{CF})_x \rightarrow x\,\overset{+1}{Li}F + x\,\overset{0}{C}$

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Nomenclature of the primary lithium batteries

Nomenclature by IEC
(International Electrotechnical Commission)

„**BR**“: B = Li/$(CF)_n$, R = Round

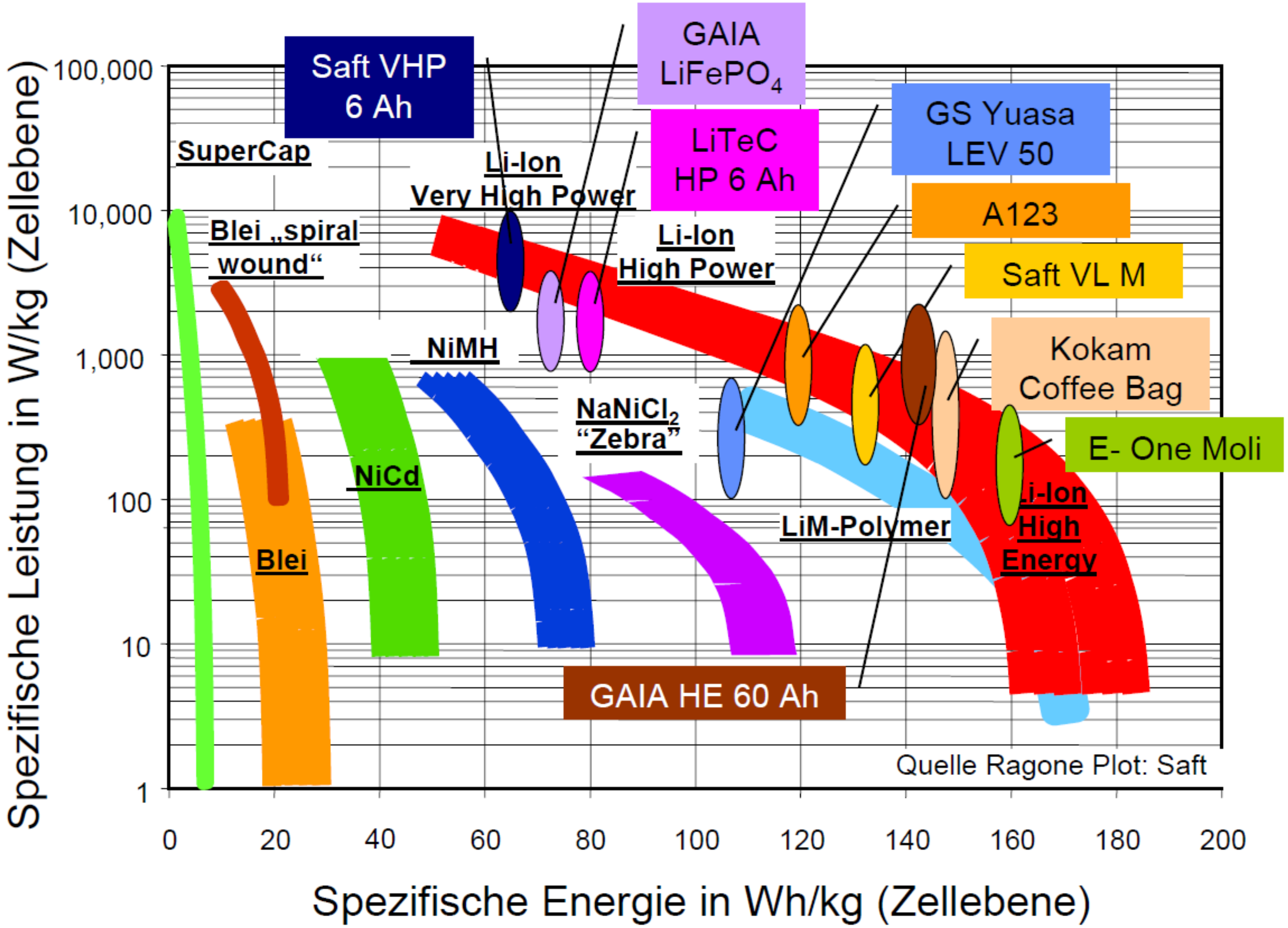
„**CR**“: C = Li/MnO_2, R = Round

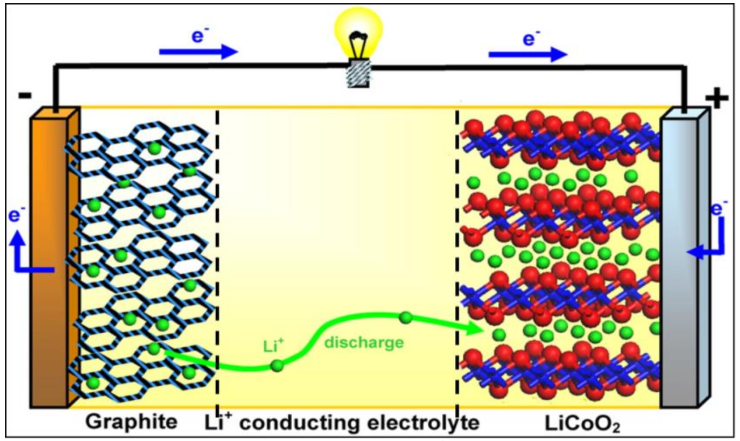
„**ER**“: E = Li/$SOCl_2$, R = Round

„**FR**“: F = Li/FeS_2, R = Round

Secondary Li-ion Systems

A „Ragone-plot“





Quelle: Bruce, Solid State Ionics 179 (2008) 752–760

The „Rocking Chair Principle"

Discharge reaction (example):

anode: $C_6Li \rightarrow C_6Li_{1-x} + x\ Li^+ + x\ e^-$ (oxidation)

cathode: $Li_yCoO_2 + x\ Li^+ + x\ e^- \rightarrow Li_{y+x}CoO_2$ (reduction)

Sum: $C_6Li + Li_yCoO_2 + x\ Li^+ + x\ e^- \rightarrow Li_{y+x}CoO_2 + C_6Li_{1-x}$

(Sum: $C_6Li + 2\ Li_{0,5}CoO_2 \rightarrow 2\ LiCoO_2 + C_6$)

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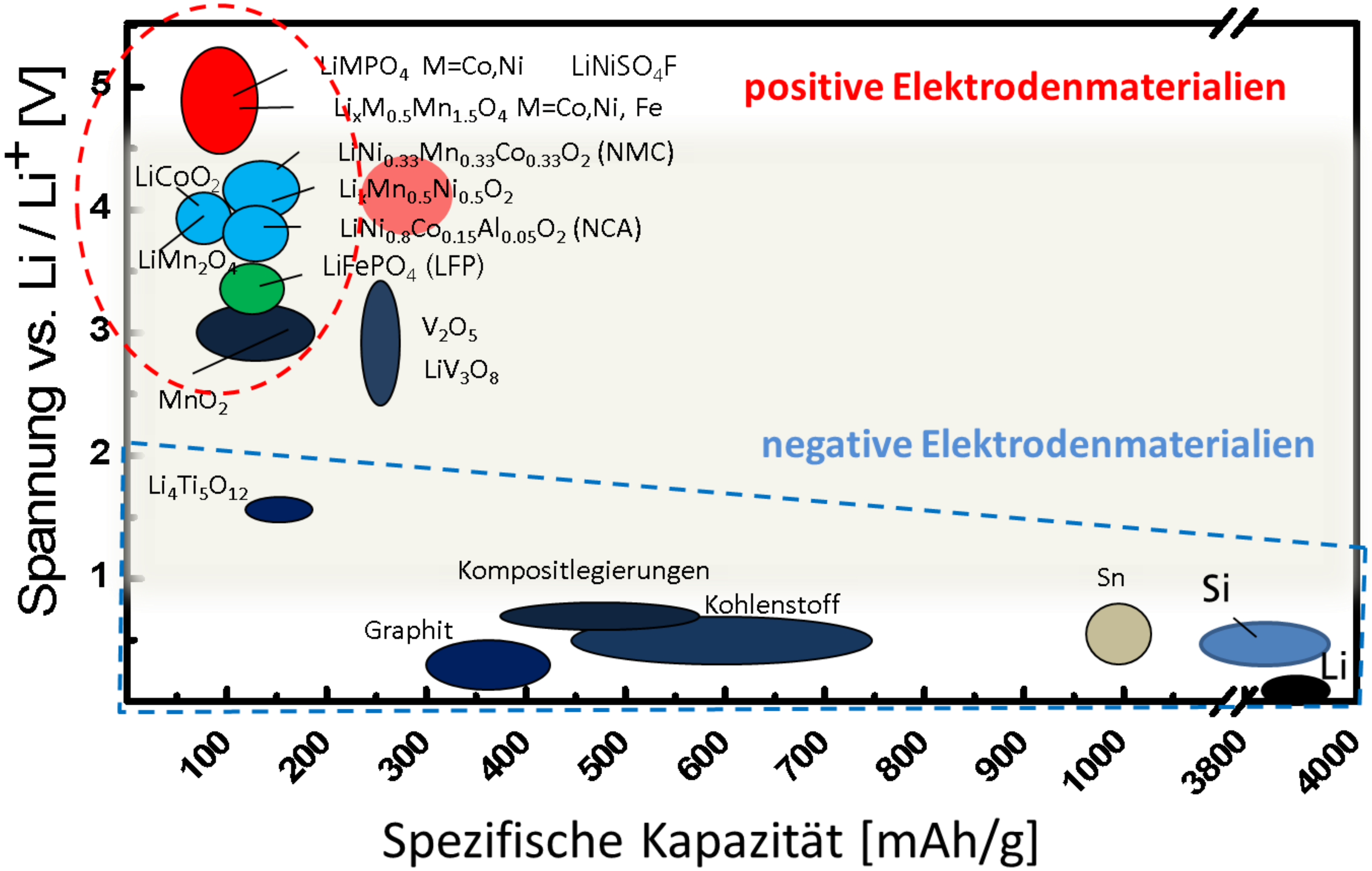
Cathode and anode materials in Li-ion batteries



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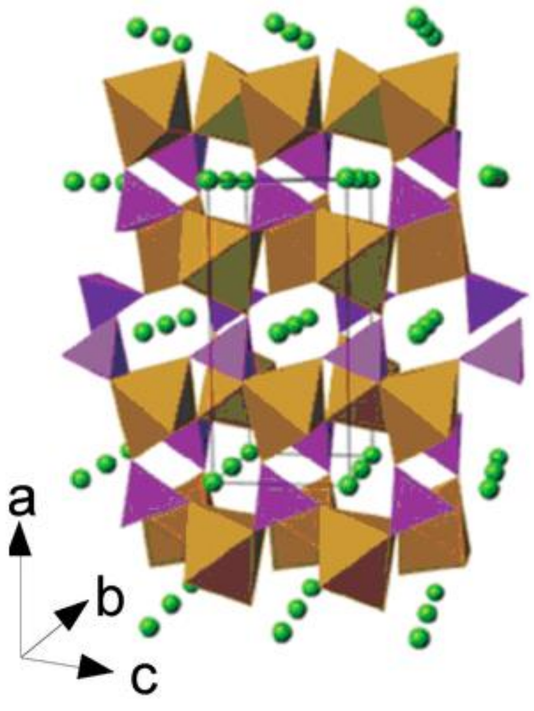
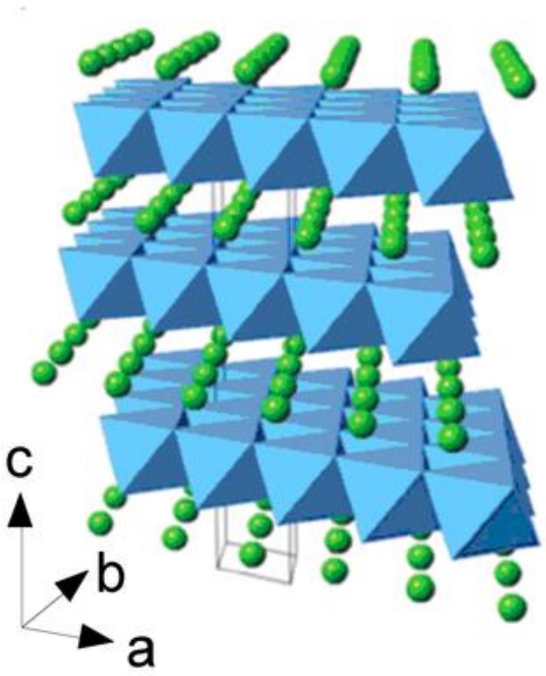
Olivine-structure



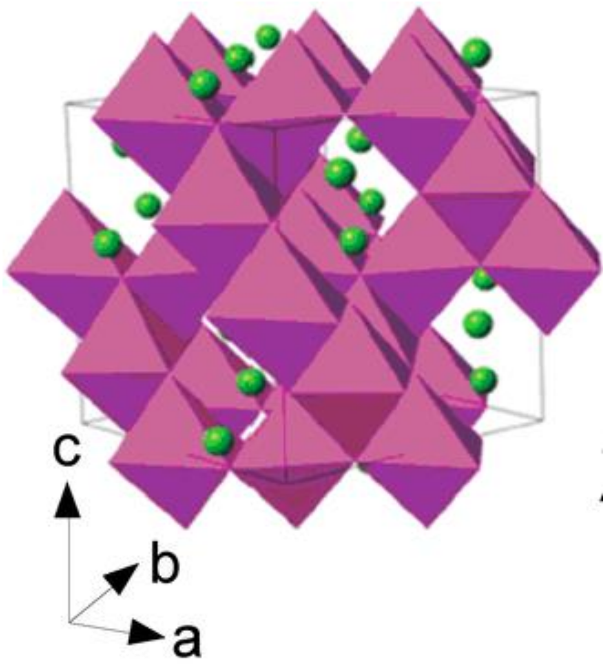
Olivine $LiFePO_4$ (1D)

Layered-structure



Layered oxide $LiCoO_2$ (2D)

Spinel-structure



Cubic spinel $LiMn_2O_4$ (3D)

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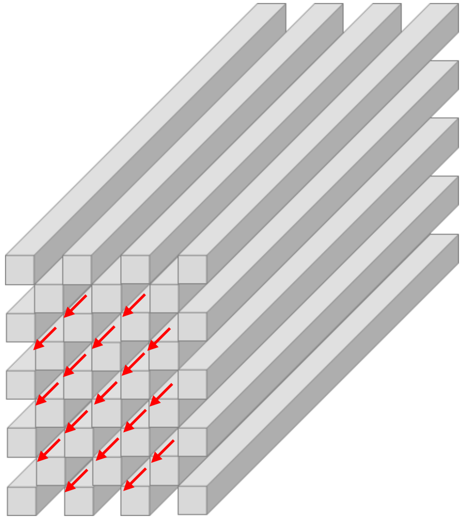
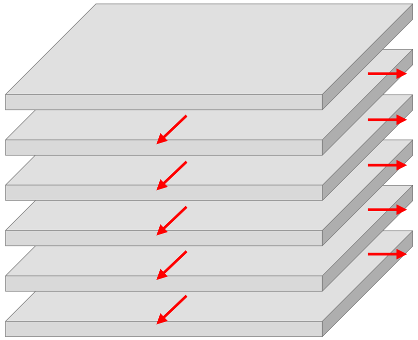
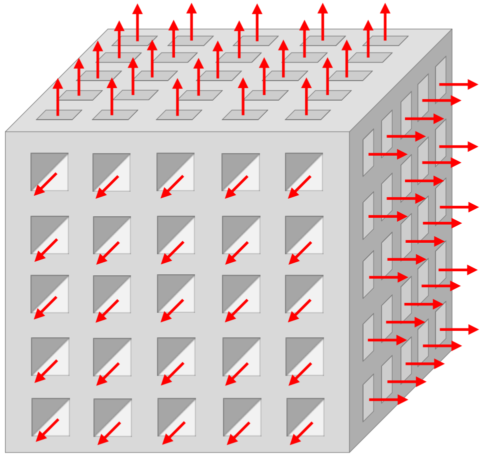
Classification of the cathode materials

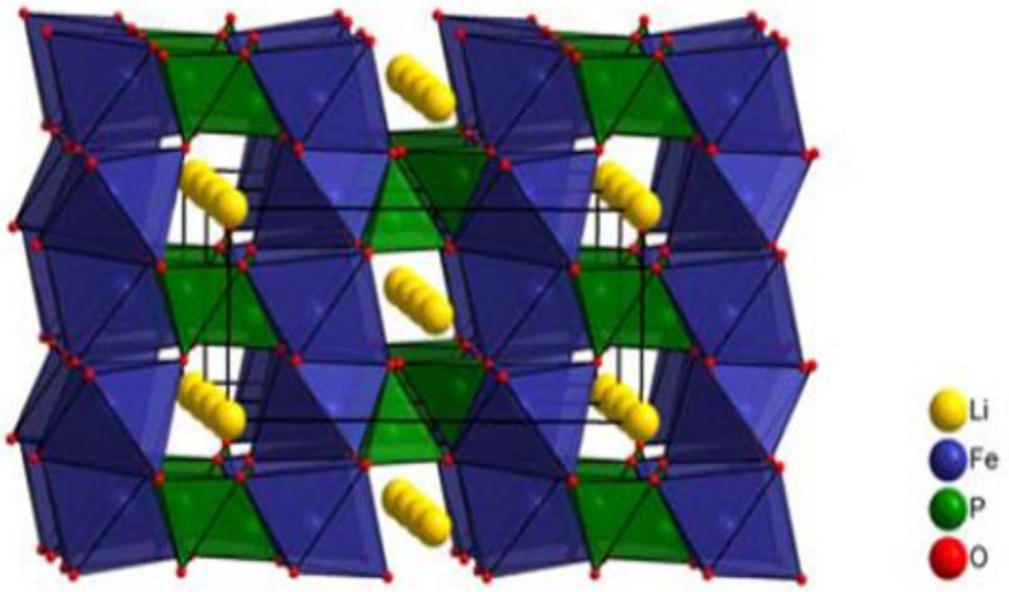
Olivine-structure	Layered-structure	Spinel-structure
		
Olivine (1D)	Layered oxide (2D)	Spinel (3D)

Dimensionality of the Li-ion transport in solids

$LiFePO_4$ - Lithium-iron(II)-phosphate (LFP)

http://www.fvee.de/fileadmin/publikationen/Workshopbaende/ws2010-1/ws2010-1_07_WohlfahrtMehrens.pdf

- ☑ environmental friendly
- ☑ cheap
- ☑ high theoretical capacity
- ☑ high stability/high safety
- ☑ „overcharge-resistant”

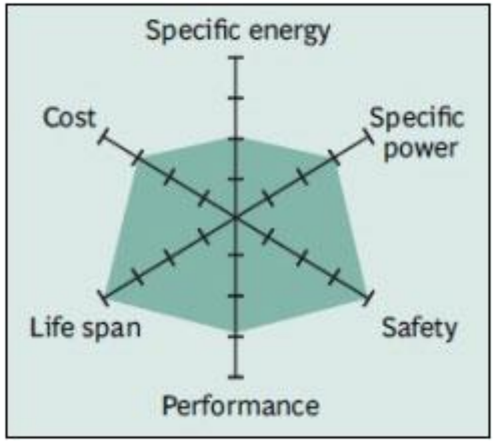


- ☒ very poor electronic and ionic conductivity

Structure

- Olivine-structure
- FeO_6 octahedrons
- PO_4 tetrahedrons

$LiFePO_4$ - Lithium-iron(II)-phosphate - summary



Lithium Iron Phosphate: LiFePO4, Graphite anode, Since 1996 Short form: LFP or Li-phosphate	
Voltage, nominal	3.20V, 3.20V
Specific energy (capacity)	90–120Wh/kg
Charge (C-rate)	1C typical; 3.65V peak; 3h charge time
Discharge (C-rate)	25-30C continuous, 2V cut-off (lower that 2V causes damage)
Cycle life	1000–2000 (related to depth of discharge, temperature)
Thermal runaway	270°C (518°F) Very safe battery even if fully charged
Applications	Portable and stationary needing high load currents and endurance
Comments	Very flat voltage discharge curve but low capacity. One of safest Li-Ions. Elevated self-discharge

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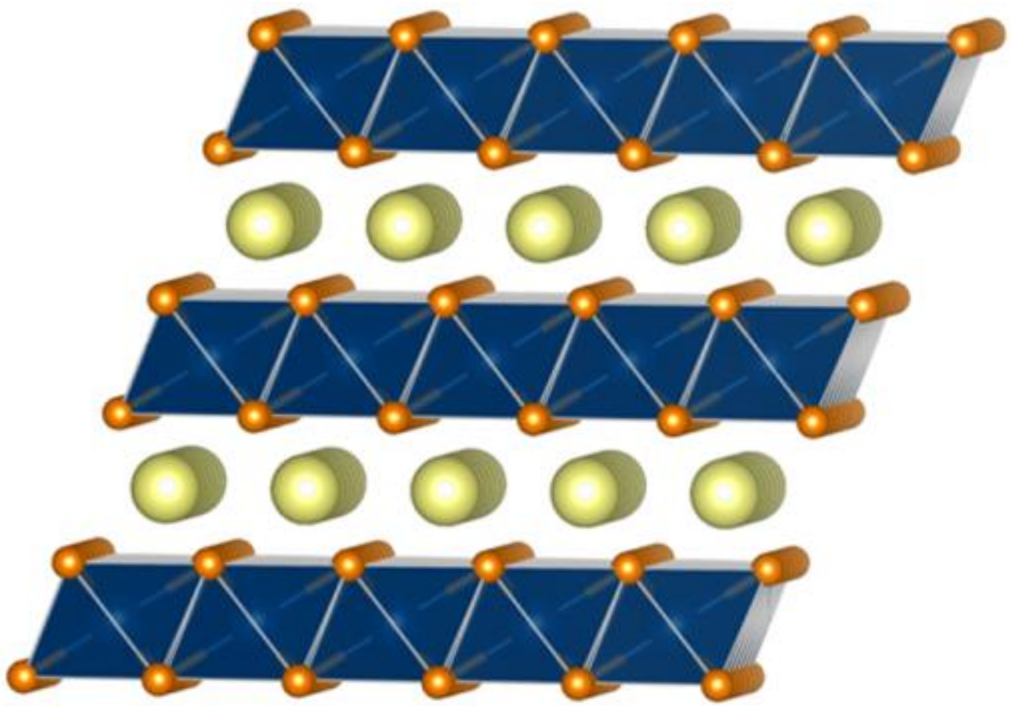
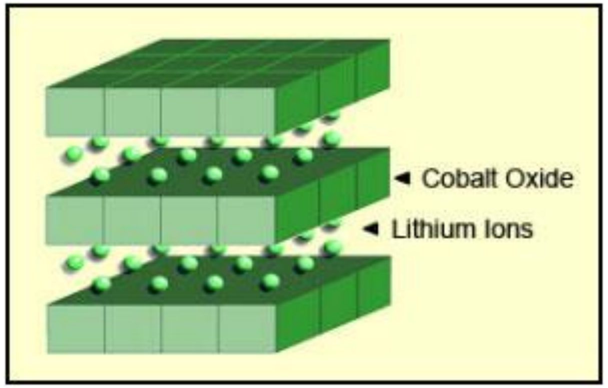
$LiCoO_2$ - Lithium-cobalt(III)-oxide (LCO)





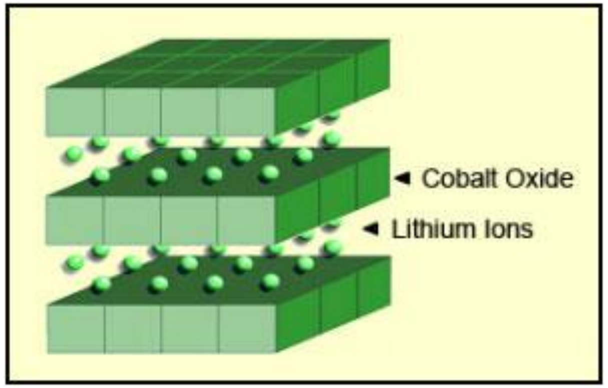
B.C. Melot, L.-M. Tarascon, *Acc. Chem. Res.*, **2012,** 46, 1227

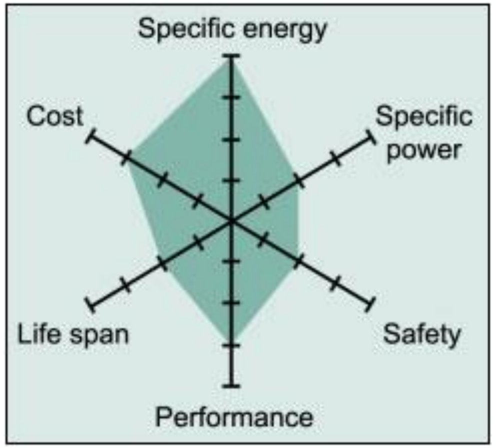
$LiCoO_2$ - Lithium-cobalt(III)-oxide

- ✓ very high theoretical capacity (ca. 274 mAh/g)
- ✓ high energy density material
- ✓ lightweight material

- ✗ High toxicity caused by cobalt
- ✗ Non-environmental friendly, harmful
- ✗ small reversible capacity (130 mAh/g)
- ✗ high costs (see price of cobalt)

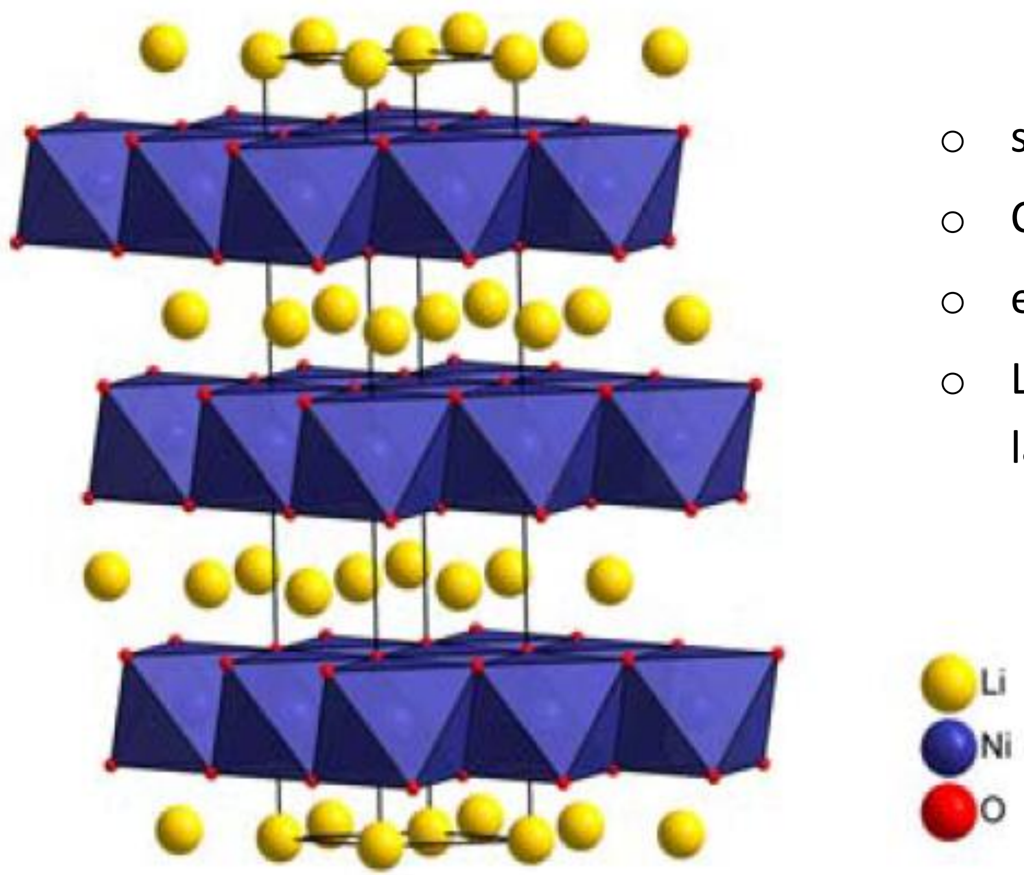
$LiCoO_2$ - Lithium-cobalt(III)-oxide - summary





Lithium Cobalt Oxide: $LiCoO_2$ (~60% Co). Graphite anode, Since 1991 Short form: LCO or Li-cobalt.	
Voltage, nominal	3.60V
Specific energy (capacity)	150–250Wh/kg
Charge (C-rate)	0.8C, 1C maximum, 4.20V peak (most cells); 3h charge typical
Discharge (C-rate)	1C; 2.50V cut off
Cycle life	500–1000, related to depth of discharge, load, temperature
Thermal runaway	150°C (302°F). Full charge promotes thermal runaway
Applications	Mobile phones, tablets, laptops, cameras
Comments	Very high specific energy, limited specific power. Cobalt is expensive. Serves as Energy Cell.

$LiNiO_2$ - Lithium-nickel(III)-oxide (LNO)



http://www.fvee.de/fileadmin/publikationen/Workshopbaende/ws2010-1/ws2010-1_07_WohlfahrtMehrens.pdf

Structure

- similar to $LiCoO_2$
- Ccp der O^{2-}
- edge-sharing NiO_6-octahedrons
- Li-ions intercalate between the layers

$LiNiO_2$ - Lithium-nickel(III)-oxide

- ☑ less toxic compared to $LiCoO_2$
- ☑ cheaper than $LiCoO_2$
- ☑ higher reversible capacity, > 150 mAh/g
- ☑ high energy density

- ☒ difficult preparation process (i.e., Ni^{3+})
- ☒ poor chemical stability
- ☒ higher safety risk

$LiNiO_2$ - Lithium-nickel(III)-oxide

The source of the poor chemical stability

$LiNiO_2$ is stable in air and also at higher temperatures

+3
$LiNiO_2$ → stable

Problems in use in the battery cell

on charging process:

- deintercalation of Li^+-ions → $Li_{1-x}NiO_2$

X ≤ 1 → oxidation number changes (+3 → +4)

+3/+4
$Li_{1-x}NiO_2$

- Ni^{4+} is non-stable → strong oxidation agent

$LiNiO_2$ - Lithium-nickel(III)-oxide

The result: internal redox reaction occurs!

Ni^{4+} oxidizing O^{2-} ions ➔ release of oxygen gas

$$2\ Ni^{4+} + 2\ e^{-} \rightarrow 2\ Ni^{3+}$$ (**Red**uction)

$$2\ O^{2-} \rightarrow O_2 + 2\ e^{-}$$ (**Ox**idation)

Strong exothermic reaction!

➔ Release of large amount of energy in form of **heat**

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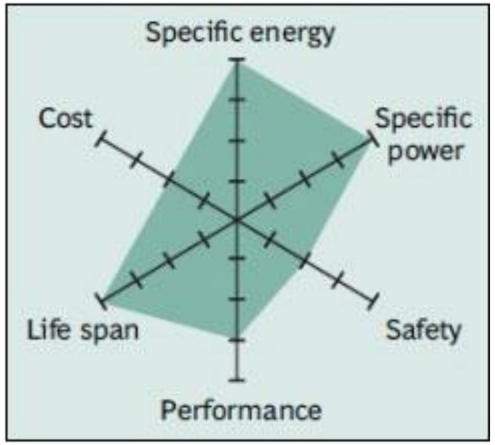
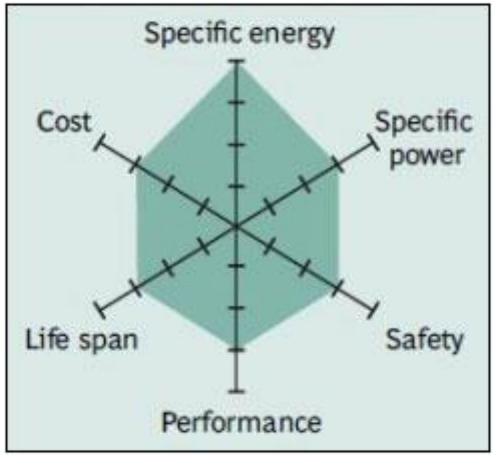
$LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ - Lithium-nickel-cobalt-aluminium-oxide (NCA)



Lithium Nickel Cobalt Aluminum Oxide: $LiNiCoAlO_2$ (~9% Co) Graphite anode Since 1999 Short form: NCA or Li-aluminum.	
Voltage, nominal	3.60V
Specific energy (capacity)	200-250Wh/kg
Charge (C-rate)	0.5C standard; 4.20V peak (most cells), 3h charge typical
Discharge (C-rate)	1C continuous; 3.00V cut-off
Cycle life	500 (related to depth of discharge, temperature)
Thermal runaway	150°C (302°F) typical, High charge promotes thermal runaway
Applications	Medical devices, industrial, electric powertrain (Tesla)
Comments	Shares similarities with Li-cobalt. Serves as Energy Cell.

$LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ - Lithium-nickel-manganese-cobalt-oxide (NMC)



Lithium Nickel Manganese Cobalt Oxide: $LiNiMnCoO_2$. Graphite anode Since 2008 Short form: NMC (NCM, CMN, CNM, MNC, MCN are similar with different medal combination)	
Voltage, nominal	3.60V, 3.70V
Specific energy (capacity)	150–220Wh/kg
Charge (C-rate)	0.7C, 4.20V peak; 3h charge time
Discharge (C-rate)	2C continuous; 2.50V cut-off
Cycle life	1000–2000 (related to depth of discharge, temperature)
Thermal runaway	210°C (410°F) typical. High charge promotes thermal runaway
Applications	E-bikes, medical devices, EVs, industrial
Comments	Provides high capacity and high power. Serves as Hybrid Cell. This chemistry is often used to enhance Li-manganese.

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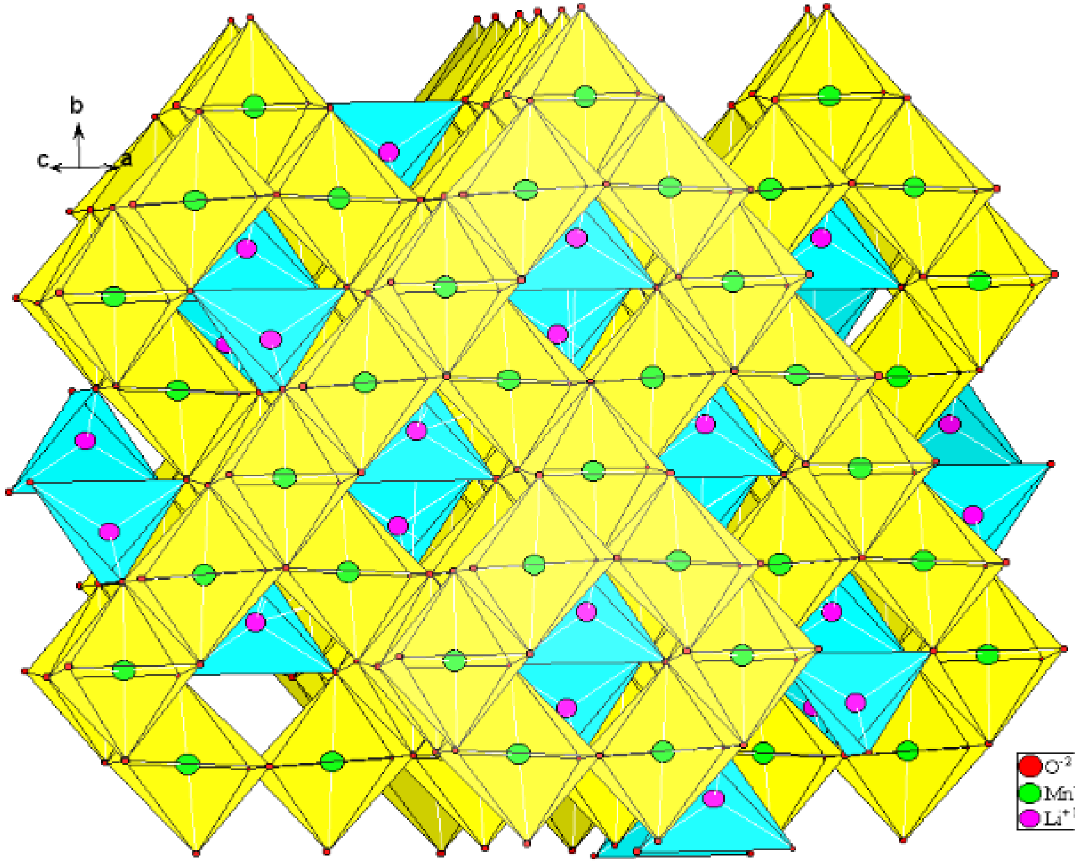
$LiMn_2O_4$ - Lithium-manganese(III/IV) oxide (LMO)



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$LiMn_2O_4$ - Lithium-manganese-oxide

- ☑ less toxic
- ☑ higher thermal stability
- ☑ cost-efficient
- ☑ Mn is a frequent element (0,95%)

- ☒ smaller reversible capacity (120 mAh/g)
- ☒ poor chemical stability

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$LiMn_2O_4$ - Lithium-manganese-oxide

Problem: poor chemical stability

- $Li_xMn_2O_4$
- changing the oxidation state of Mn by variation of *x*

x	Compound	Oxidation number of manganese ions
1	$Li_1Mn_2O_4$	+3,5
2	$Li_2Mn_2O_4$	+3
0	$Li_0Mn_2O_4$	+4

Disproportionation von Mn^{+3}

$$2\,Mn^{3+} \longrightarrow Mn^{4+} + Mn^{2+}$$

Mn^{4+} ↓ solid

Mn^{2+} ↓ soluble in the electrolyte

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$LiMn_2O_4$ - Lithium-manganese-oxide

Problem: poor chemical stability

Transport of Mn^{2+} to the anode

⬇

Deposition

⬇

Oxidation of Li by Mn^{2+}

$Mn^{2+} + 2\,e^- \rightarrow Mn$ (Reduction)

$2\,Li \rightarrow 2\,Li^+ + 2\,e^-$ (Oxidation)

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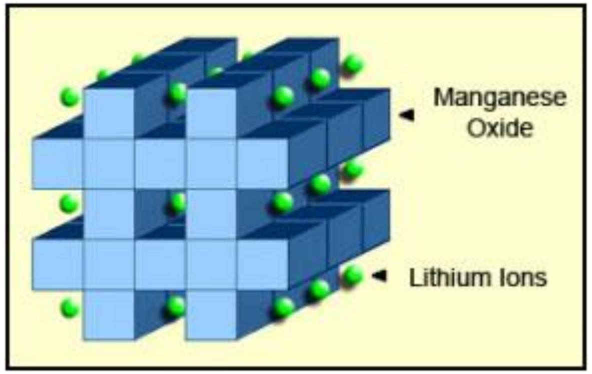
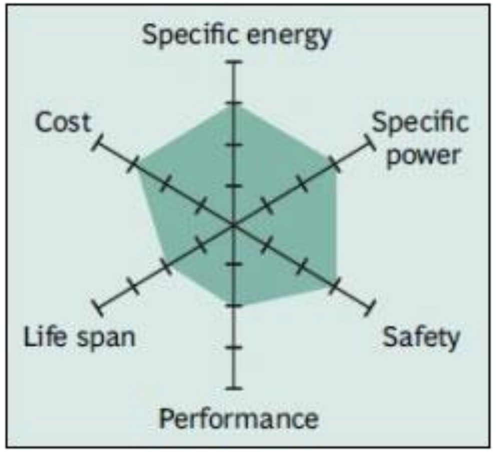
$LiMn_2O_4$ - Lithium-manganese-oxide - summary





Lithium Manganese Oxide: $LiMn_2O_4$, Graphite anode, Since 1996 Short form: LMO or Li-manganese (spinel structure)	
Voltage, nominal	3.70V (some may be rated 3.80V)
Specific energy (capacity)	100–150Wh/kg
Charge (C-rate)	0.7–1C recommended, 3C maximum; 4.20V peak (most cells)
Discharge (C-rate)	10C continuous, 30C for 5s pulse, 2.50V cut-off
Cycle life	500–1000 (related to depth of discharge, temperature)
Thermal runaway	250°C (482°F) typical. High charge promotes thermal runaway
Applications	Power tools, medical devices, electric powertrains
Comments	High power but less capacity; safer than Li-cobalt; commonly mixed with NMC to improve performance.

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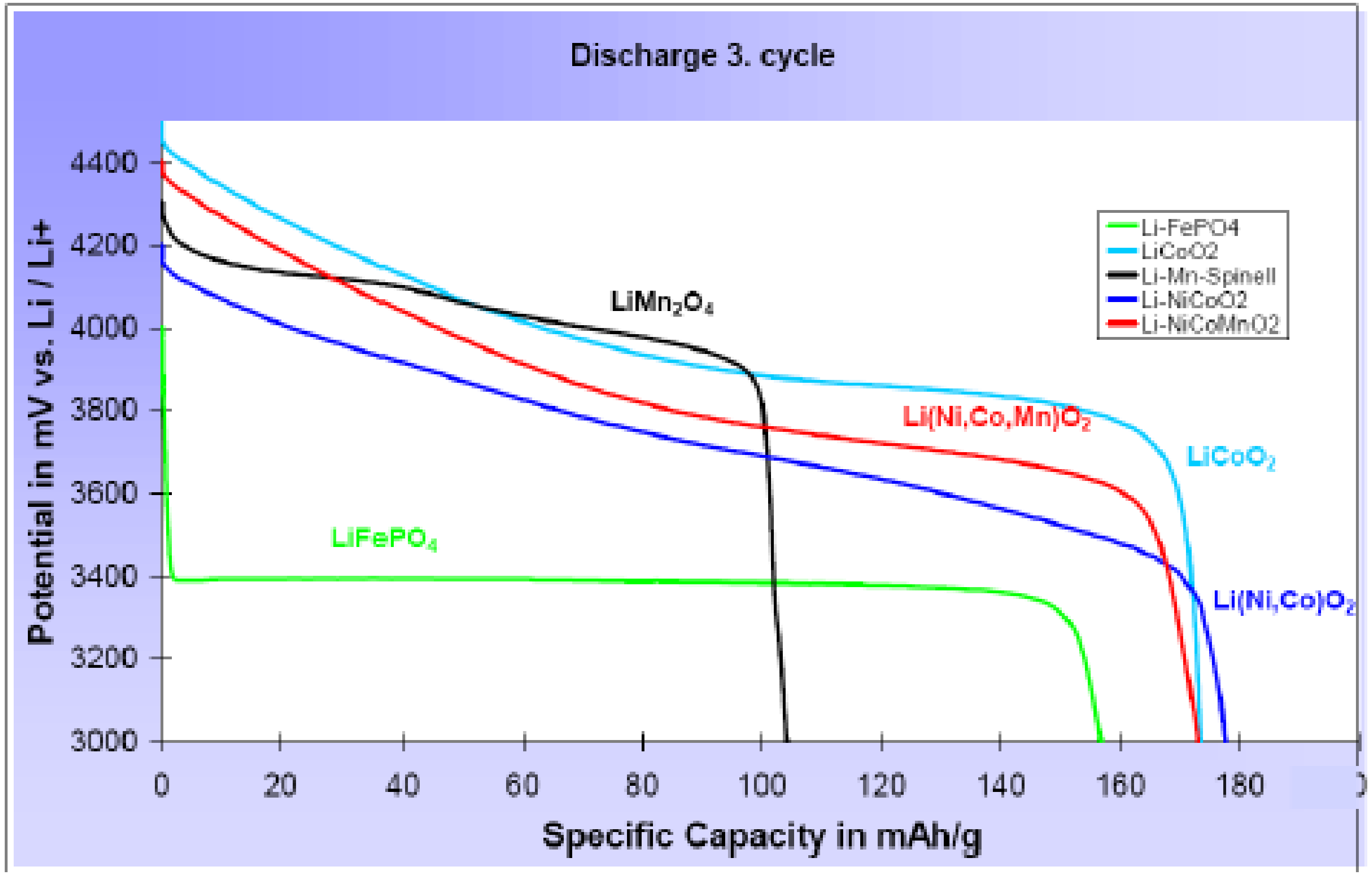
Cathode materials - summary

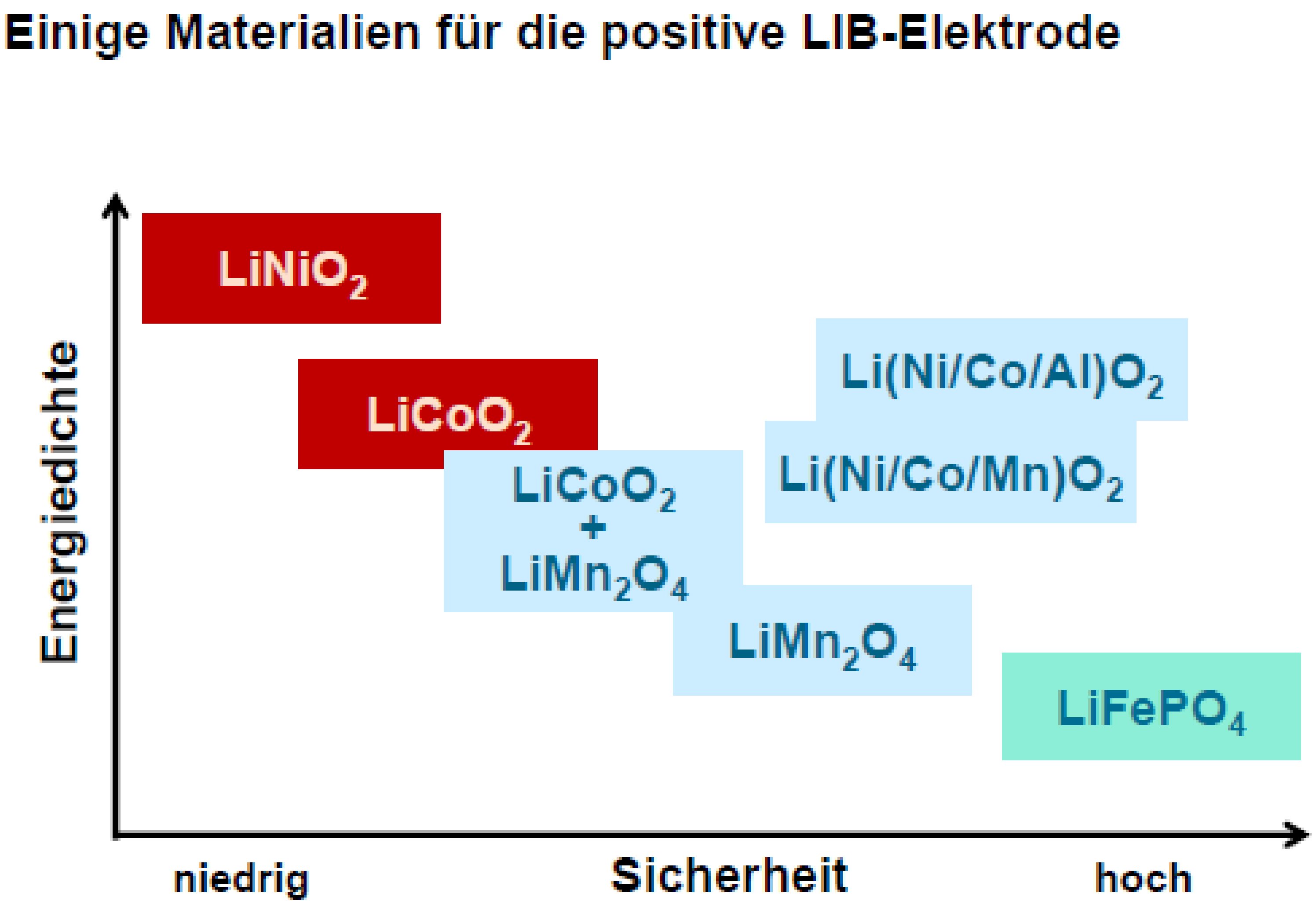
Material	Spannung	Spezifische Kapazität	Spezifische Energie
$LiCoO_2$	3,7 V	140 mAh/g	0,518 kWh/kg
$LiNiO_2$	3,5 V	180 mAh/g	0,630 kWh/kg
$LiCo_{1/3}Ni_{1/3}Mn_{1/3}O_2$	3,6 V	160 mAh/g	0,576 kWh/kg
$Li(Li_aNi_xMn_yCo_z)O_2$	4,2 V	220 mAh/g	0,920 kWh/kg
$LiMn_2O_4$	4,0 V	100 mAh/g	0,400 kWh/kg
$LiFePO_4$	3,3 V	150 mAh/g	0,495 kWh/kg
Li_2FePO_4F	3,6 V	115 mAh/g	0,414 kWh/kg

Different performance, costs and environmental impact

Unterschiedliche Potentiallagen



Cathode materials - summary



Construction of Li-ion batteries

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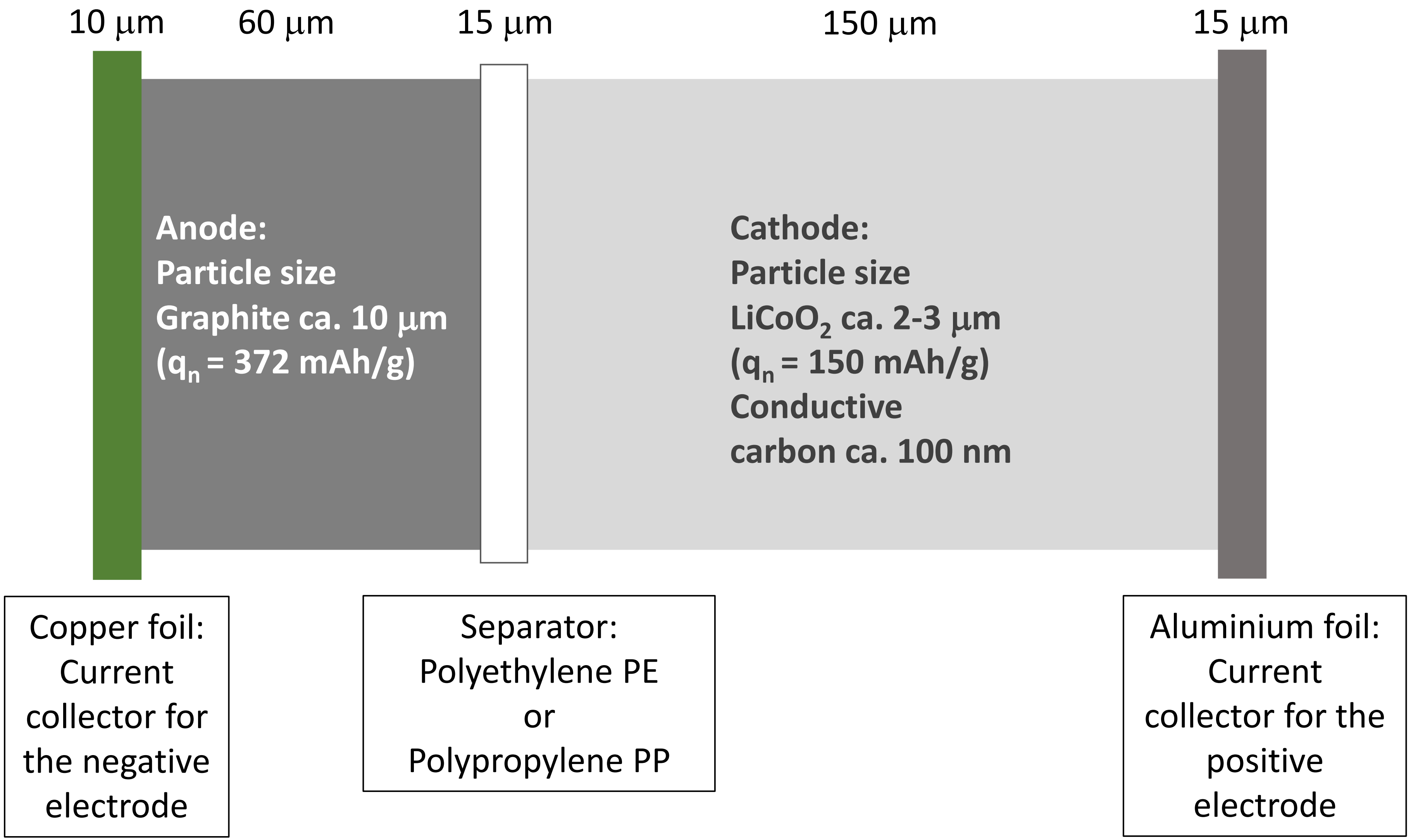
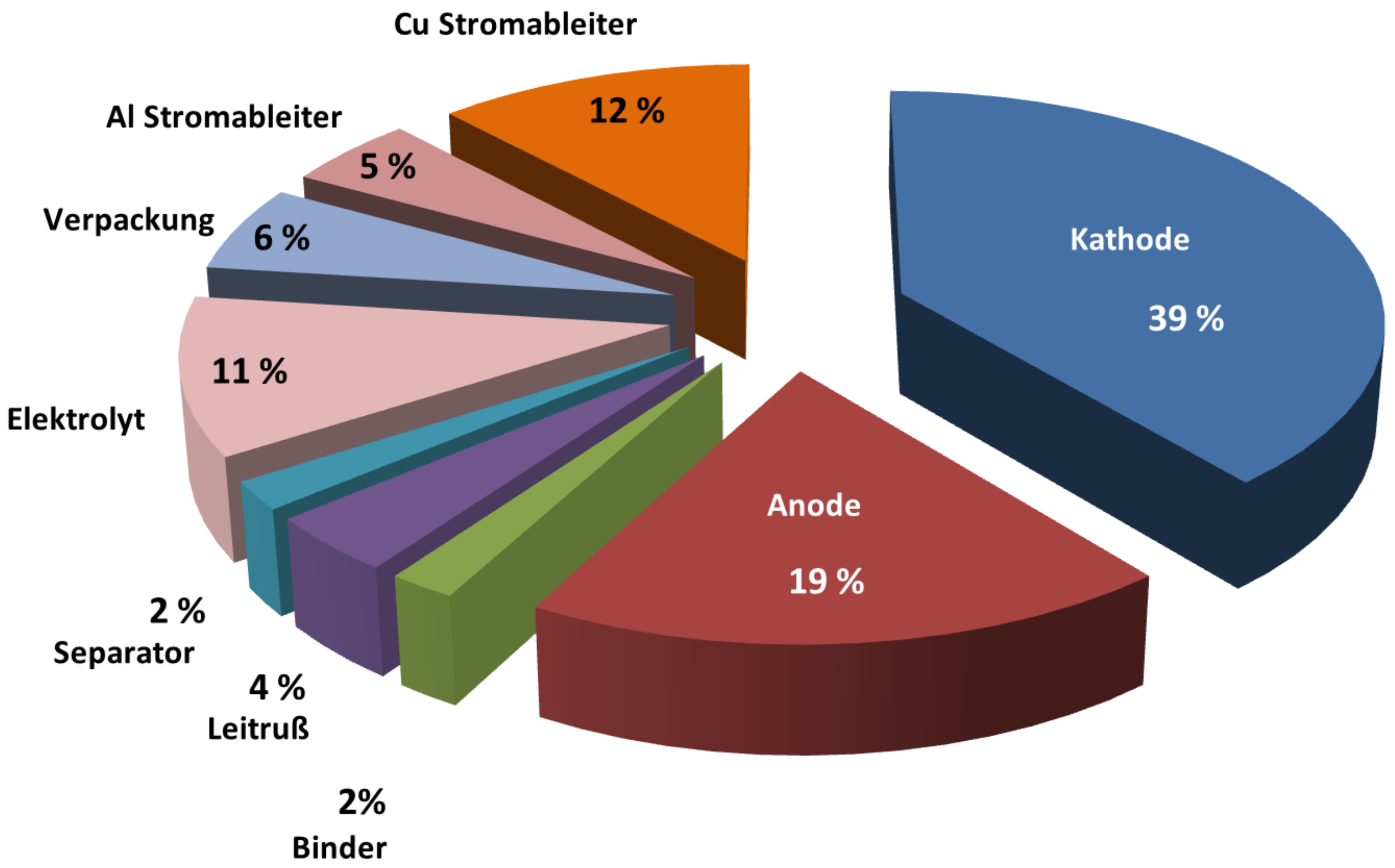
Construction of the Li-ion batteries (proportions)

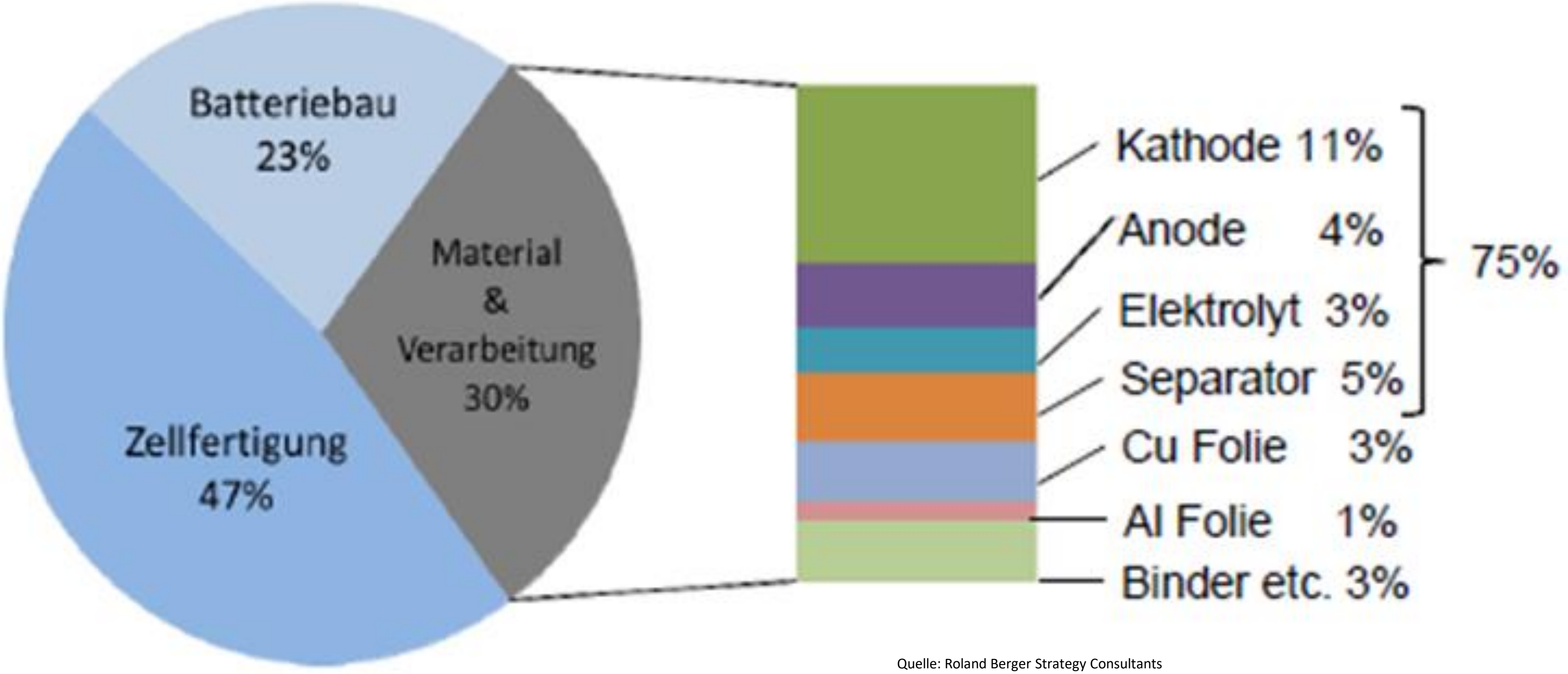


Weight distribution of the elementary components of a Li-ion battery cell



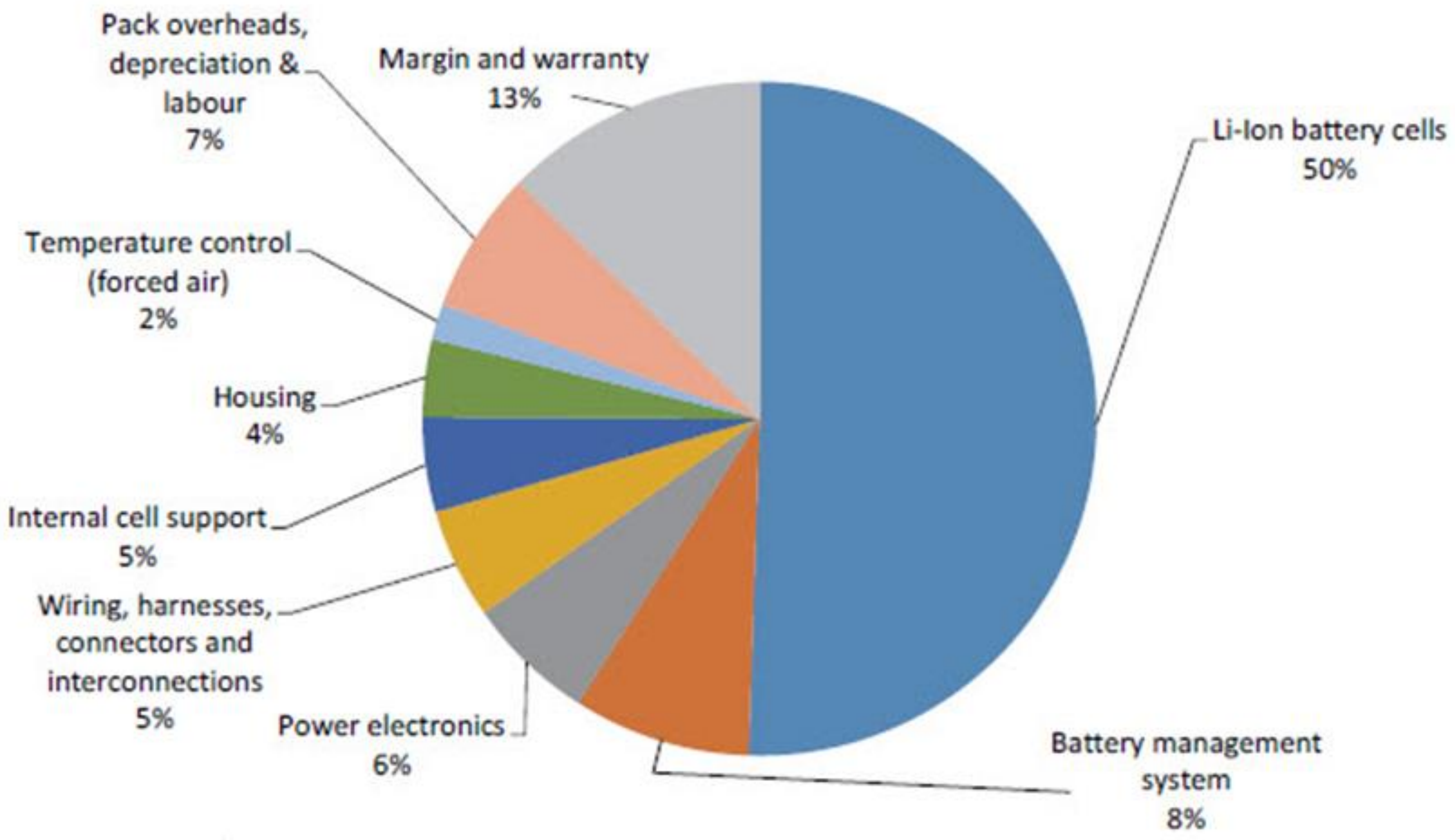
Cost distribution of a Li-ion battery pack

Based on a 500€/kWh high energy pack



Quelle: Roland Berger Strategy Consultants

Cost distribution of a 22 kWh Li-ion battery pack used in a mid-size full-EV (2012)

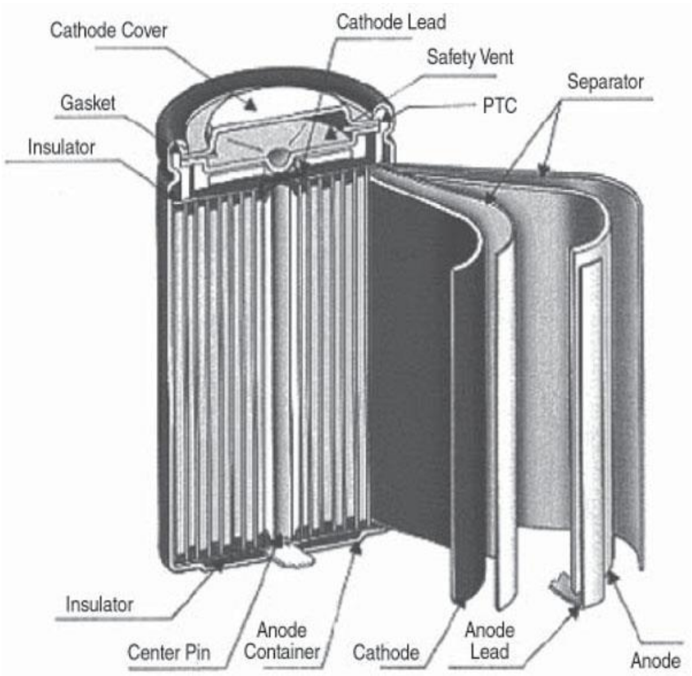


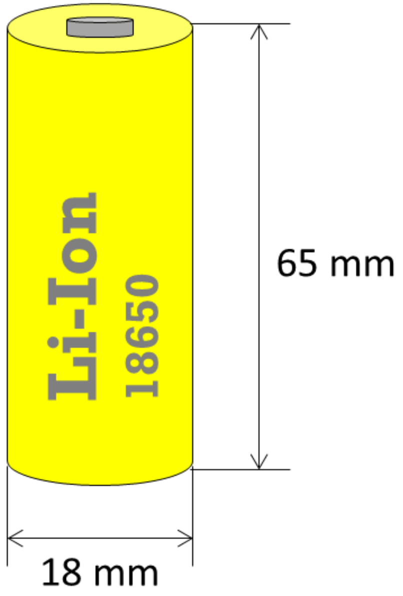
$500-800/kWh - Pack
$300-400/kWh - Zelle

Quelle: Element Energy, 2012

Custom Li-ion battery cell formats

Cylindrical cell





Typ 18650

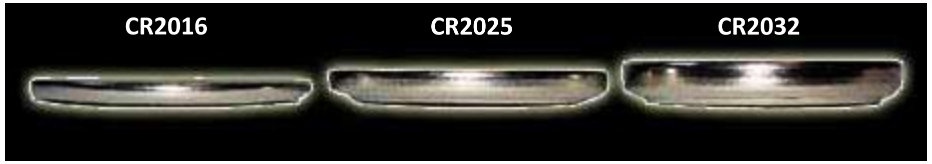
z.B. 3S3P; 9 x 3,6V @ 2400mAh Zelle = 10,8V @ 00 mAh

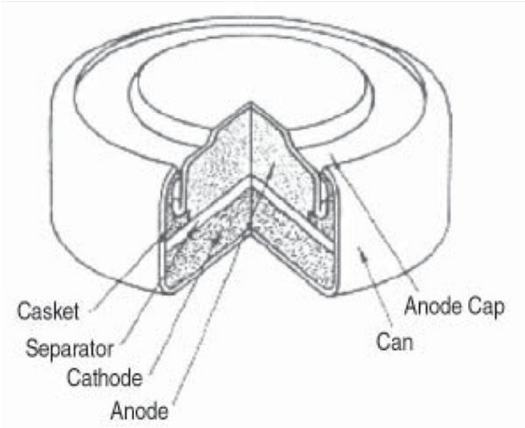
Typ 18650

Tesla Model S Batterie: >7000 individuelle 18650 Zellen in 16 Modulen.
85 kWh (400V DC)

Custom Li-ion battery cell formats

Button cell (primary cell)

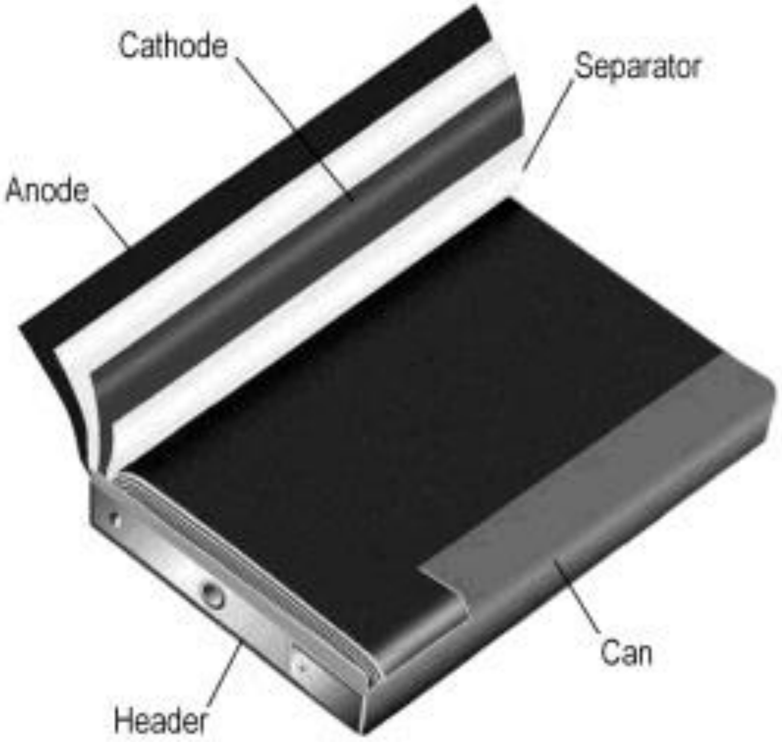




Bezeichnung	Durchmesser (mm)	Höhe (mm)	Spannung (V)	Kapazität (mAh)
CR2016	20	1,6	3	90
CR2025	20	2,5	3	150
CR2025	20	2,5	3	165
CR2032	20	3,2	3	210

Custom Li-ion battery cell formats

The prismatic form



Comparison of the different cell chemistries

	Lead acid (VRLA)	NiCd	NiMH	Li-Ion
Nominal cell voltage (V)	2,0	1,2	1,2	3,7
Specific energy(Wh/kg)	35	50	90	165
Energy density (Wh/l)	80	170	330	330
cost/kWh	50	200	200	300-500
Cycle life performance	200	600-1000	300-500	500