

Li-ion batteries for electric vehicles*

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This paper presents brief outlook for Li-ion battery market for electric vehicles in comparison with that for portable electronics. The second part of the article presents electrochemical model as well as the main points of material issues in lithium batteries. Review of currently available and possible for future application cathode materials is provided. Stress is placed on importance transport properties of materials.

1. INTRODUCTION

Market of rechargeable electrochemical cells currently experience dynamic growth, which is connected with immense interest of automotive industry. This is due to planned employment of batteries as energy sources for hybrid and electric vehicles (so called Battery Electric Vehicles, BEVs). Widely commercialised Ni-Cd and NiMH-type reversible cells technology already have achieved their limits. Thus, Li-ion batteries [1] become the most promising technology of reversible cells. The majority of great automotive companies (Nissan Motor, General Motors Co., Honda Motor Co., ...) have presented wide plans of production of electric vehicles powered by Li-ion batteries for the forthcoming five years. Corporations manufacturing lithium batteries feel enormous pressure from car industry. Fast growing market of large capacity batteries for electric vehicles soon will overtake sell of lithium batteries for other applications (mobile phones, notebooks, etc.). Market of rechargeable Li-ion batteries in 2009

*This article is dedicated to Professor Dobiesław Nazimek on the occasion of his 65th birthday

reached \$8.4 billion [2]. It is estimated that 97% of it falls into portable electronics (mobile phones and notebooks), 3% – electric vehicles (\$0.25 billion). Fraction of BEVs will grow up to \$3 billion in 2010 and in 2012 will exceed value of portable electronics achieving \$15.8 billion. Its size is predicted to achieve \$22.5 billion in 2014, which means 90-fold growth in five-years time. Figure 1 shows Li-ion batteries market in 2009-2014 period.

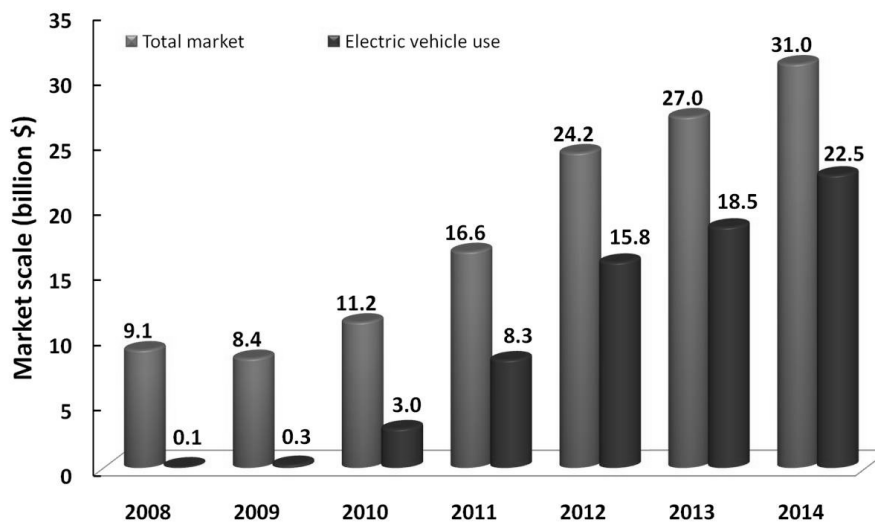


Fig. 1. Size of Li-ion batteries market in 2008-2014 [2].

Capacity required for Li-ion battery for application in electric vehicle is considerably large in comparison with those for portable electronics. For instance, capacity of the battery in a mobile phone is 2-3 Wh, whereas hybrid vehicle needs 1 kWh (500 times more) and fully electric car at least 20 kWh (10,000 times more). Figure 2 depicts difference in capacity required for portable electronics and automotive application.

It is estimated that Li-ion batteries for portable electronics accumulate total energy of 10,000 – 15,000 MWh. Assuming that global annual sale of new cars reaches 70 million and only 1% of them are BEVs and 20% are hybrid vehicles, the market size (28,000 MWh) will double that of portable electronics. If planned growth of market of Li-ion batteries for automotive industry takes place, prices of batteries may drop from current 2000 \$/kWh to 500 \$/kWh, and, as

a consequence, new applications will appear: in stationary installations, mobile and industrial devices. Figure 3 shows new possibilities of application of high-capacity Li-ion batteries.

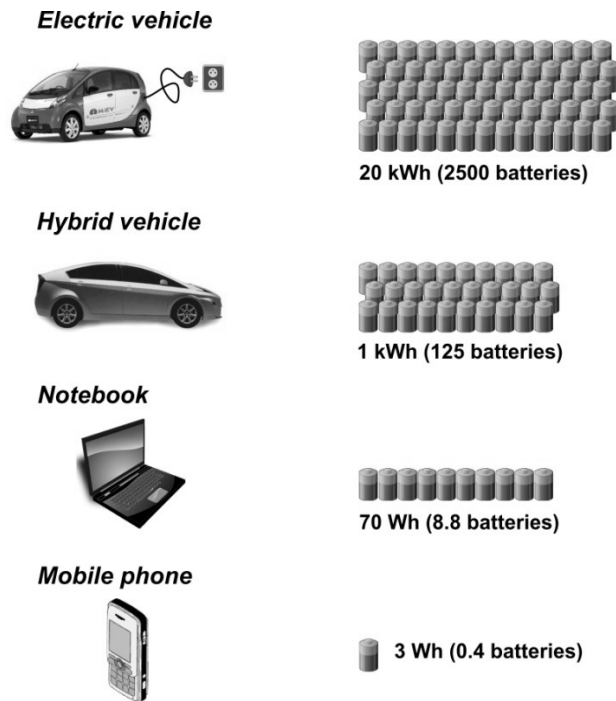


Fig. 2. Energy demand of typical devices and electric vehicles (shown as required number of 8 Wh 18650-type batteries) [2].

High capacity Li-ion batteries may play a significant role in future economy based on renewable energy sources. Energy production from solar or wind power plants is strongly dependent on unstable environmental conditions, which in turn cause instability of energy supply and difficulties with connection to a large scale power network, as well as fluctuations of current frequency and reduction of power quality. If power generated from renewable energy sources is stored in high-capacity Li-ion batteries, it could be transferred to power grid maintaining steady performance. In such way power plants, buildings and factories could be powered.

In 2008–2009 development of Li-ion batteries technology evidently gain pace. In the USA, China, Japan, South Korea and France dedicated national programmes were established (USA – \$2.4 billion, France – \$2.2 billion, China – 863 programmes, Germany €0.5 billion). Two Japanese corporations (Sanyo

Electric Co., Panasonic Corp.) alone will have invested \$3 billion by 2012. South Korea and China are also active in this field (\$1.5 and 3 billion, respectively). China is developing its own Li-ion batteries market for at least 20 million electric motorbikes and motor scooters along with 12 million electric cars.

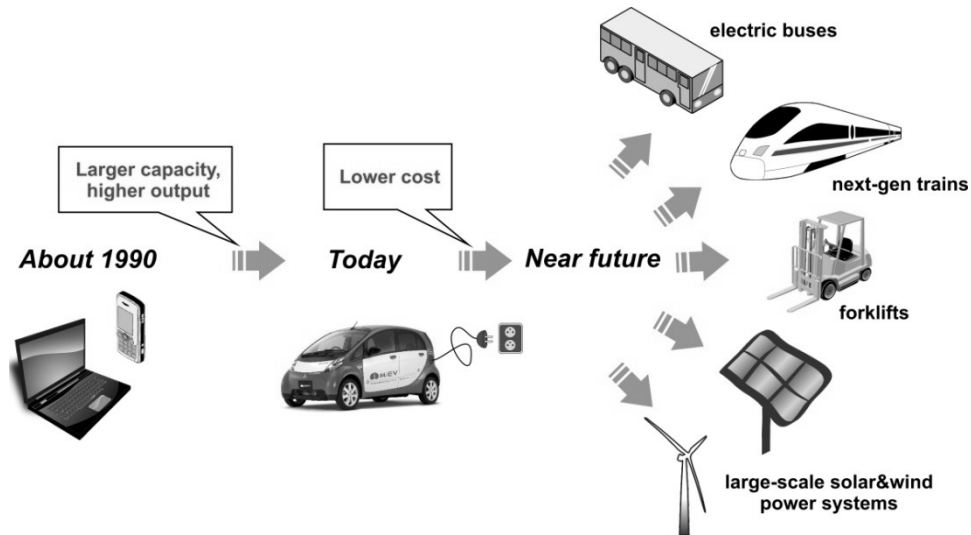


Fig. 3. New possibilities of application of high-capacity Li-ion batteries [2].

Currently there are 84 global high power lithium battery manufacturers, from which 71 are located in Far East (China, Japan, Taiwan, South Korea, Malaysia), 12 in the USA and Canada and 1 in Germany (Figure 4).

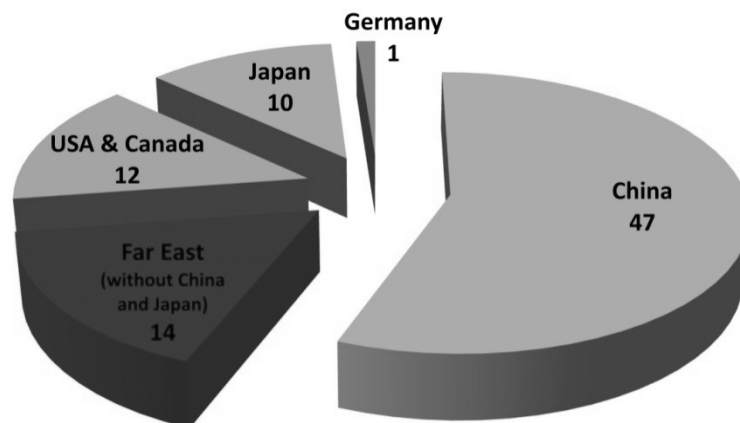


Fig. 4. Li-ion batteries global manufacturers.

The main problem in wide implementation of Li-ion batteries in vehicles lays in high cost of production in 2010–2015 period when transition from small to large scale market in will take place. Planned fast development of Li-ion batteries market for application in automotive industry requires investments in equipment and assembly lines, and, what is more, imposes strict demands for component materials for the cells. Huge funds are invested in elaboration of new cathode materials, which make 30% of the whole cell cost, of performance suitable for automotive application. The issue is lack of data concerning actual lifetime of Li-ion batteries exploited in changing environmental conditions – required battery lifetime in a car is over 10 years, whereas for notebooks 1-4 years. Safety of application in vehicles is at least of equal importance. Possessing higher capacity than battery for portable electronics, they gather more energy and, therefore, safety issues are even more strict. Cathode materials showing less reactivity along with electrolytes with higher chemical and thermal stability are sought. Additional separators covered with inert ceramic layer and placed at the electrode/electrolyte interfaces. Furthermore the matter of heat flow within the cell compartment and casing is vital. Lifetime, dependent on depth of discharge in work cycles, will be more challenging issue, even more difficult in hybrid than in BEVs. Also high (40°C) and low (-40°C) temperatures of exploitation will have deteriorating effect on battery lifetime.

2. LI-ION BATTERIES - WORKING PRINCIPLE AND WAYS OF IMPROVEMENT

Li-ion batteries employ ability of layered and tunnel-structured transition metal compounds M_aX_b (M – transition metal; X = O, S) to reversible lithiation [1,3]. In some cases it is possible to introduce more than one mole of lithium per one mole of M_aX_b at room temperature without significant changes in crystal structure, except small reversible evolution of lattice parameters. Stability of crystal structure is correlated with stability of strong ionic-covalent chemical bonds between M and X atoms. The main types of structure able to intercalate lithium are shown in Figure 5.

Intercalation of lithium into transition metal compounds M_aX_b , which is always a process involving ion and electron transfer (together with Li^+ ions equivalent number of electrons is incorporated), can be written as:



This process uses the energy of deep levels of d-type electron of transition metal (a few eV/atom), which creates possibility to accumulate energy of a few

kWh/kg and build energy storage devices of high volumetric and gravimetric energy density.

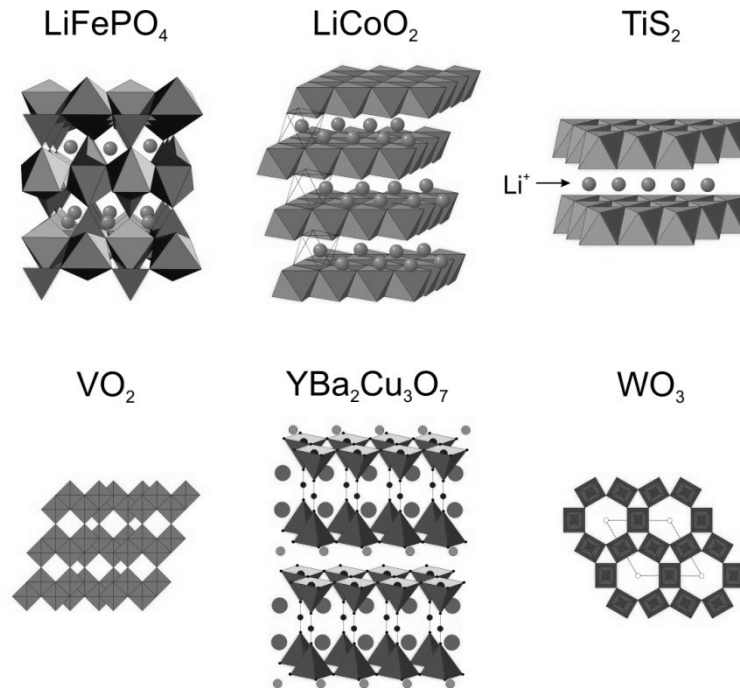


Fig. 5. Types of structure able to intercalate lithium.

Mechanism of intercalation in a $\text{Li} / \text{Li}^+ / \text{Li}_x\text{M}_a\text{X}_b$ cell is described in the literature on basis of the crystallographic-thermodynamic model [4], which was built on the model of chemical absorption. Within this model attempts to explain diversified character of discharge curve of a cell with $\text{Li}_x\text{M}_a\text{X}_b$ cathode based on crystal structure, creation of superstructures etc. are not satisfying and do not lead to formulation of a selection criterion for high performance materials.

Long-lasting author's research on various $\text{Li}_x\text{M}_a\text{X}_b$ systems: Li_xTiS_2 [5], Li_xCoO_2 [6], LiNiO_2 [7], $\text{Li}_x(\text{Co,Ni,Mn})\text{O}_2$ [8], Li_xVO_2 [9], Li_xNbSe_2 [10], Li_xWO_3 [11], $\text{Li}_x\text{YBa}_2\text{Cu}_3\text{O}_7$ [12], $\text{Li}_x\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ [13], $\text{Li}_x\text{Mn}_2\text{O}_4$ [14], Li-graphite [15], Li_xFePO_4 [16] showed that lithium intercalation into transition metal compounds with metallic or semiconducting properties is a topotactic redox reaction, in which transition metal ion changes its oxidation number, thus electron structure plays significant role in this process. Electrons (together with equivalent number of lithium ions) introduced into cathode material locate at available electron levels and raise the Fermi level in a way, which depends on

density of states function. Figure 6 shows electron scheme of $\text{Li}/\text{Li}^+/\text{Li}_x\text{M}_a\text{X}_b$ cell and depicts difference of chemical potentials of electrons in cathode and anode materials and related electromotive force of a cell.

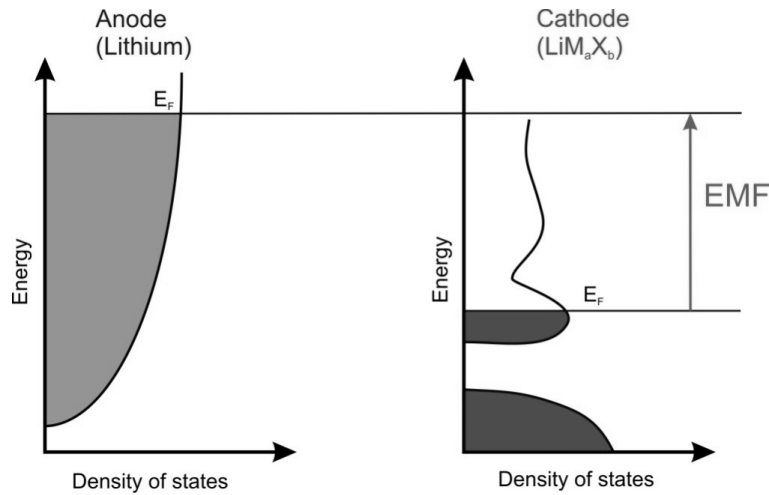


Fig. 6. Density of electron states in $\text{Li}_x\text{M}_a\text{X}_b$ and lithium depicting difference of chemical potentials of electrons and related electromotive force of $\text{Li}/\text{Li}^+/\text{Li}_x\text{M}_a\text{X}_b$ cell.

Variation of electromotive force of $\text{Li}/\text{Li}^+/\text{Li}_x\text{M}_a\text{X}_b$ cell is dependent solely on variation of chemical potential of electrons (the Fermi level) in cathode material. This is owing to constant potential of anode (constant concentration of Li^+ in electrolyte) and minute changes of chemical potential of lithium ions in cathode material (of the order of $k_B T$, which is 0.025 eV at room temperature) [17]. Variation of chemical potential of electrons in the cathode can be of the order of band width i.e. 1 eV or more. Such approach to the process of lithium intercalation allows to predict and design performance of cathode material on the basis of its electronic properties. High, monotonic density of states at the Fermi level in a cathode material and their delocalization leads to weak (advantageous from application point of view) dependence of cathode potential as a function of composition x_{Li} , wide range of lithium nonstoichiometry (high capacity) and high current density due to fast relaxation of a cathode material. On the contrary, non-continuous function of density of states $N(E)$ and localized electron states at the Fermi level is a kinetic barrier for the intercalation process, which strongly limits current density and do not allow to employ whole cathode material's capacity. Thus, nature of electron states at the Fermi level is of extreme importance for the process of intercalation, since charge transfer occurs at the Fermi level.

Presented above discussion shows that research on changes of $\text{Li}^+/\text{Li}_x\text{M}_a\text{X}_b$ potential by measurement of electromotive force of $\text{Li}/\text{Li}^+/\text{Li}_x\text{M}_a\text{X}_b$ cell is a powerful tool of solid state physics, which allow to directly observe changes of the Fermi level of $\text{Li}_x\text{M}_a\text{X}_b$ material during “doping” with lithium. Research on physicochemical properties of cathode material as a function of intercalation level (lithium content) has deep scientific aspect, since they allow to understand interrelation between crystal structure, electron structure, chemical composition, oxidation number of transition metal, disorder state and reactivity of solids. It is still an open question in the field of materials science.

Previously recalled cells with lithium metal anode ($\text{Li}/\text{Li}^+/\text{Li}_x\text{TiS}_2$), introduced into sale in 1970s, turned out to be dangerous and immediately were withdrawn from the market. This occurred since in the charge process lithium at the anode side was recovered in a form of dendrites (needle-like crystals), causing short-circuit and explosion. After twenty years of research Sony Corp. in 1991 introduced new generation of lithium batteries, so called Li-ion batteries, in which lithium metal anode was replaced with graphite. Graphite possesses ability to intercalate lithium reversibly and has beneficently low potential vs. lithium, which does not absorb electromotive force of the cell.

The main parameter of $\text{Li}_x\text{C}_6/\text{Li}^+/\text{Li}_{1-x}\text{M}_a\text{X}_b$ Li-ion batteries, which is energy density per mass or volume unit, connected with electromotive force of a cell and its capacity, is determined by crystal and electron structure of both electrode materials. Intercalation taking place can be written as:

at the anode side:



at the cathode side:



Current density drawn from a cell is determined by ionic-electronic transport in both electrode materials (ambipolar diffusion). Thus cell voltage, capacity, energy and current density are determined by properties of cathode and anode materials, whereas number of charge/discharge cycles and lifetime of a cell is electrode/electrolyte interface dependant. Operational safety depends on thermal and chemical stability of electrode materials and electrolyte. As it was shown [15] graphite anode does not limit performance (current density and voltage) of Li-ion battery, thus **possibility of improvement of overall performance of Li-ion battery lays in improvement of cathode material.**

Nowadays, LiCoO_2 as well as $\text{LiCo}_{1-y}\text{Ni}_y\text{O}_2$ and LiMn_2O_4 are employed in Li-ion batteries technology. However, all these materials have various

disadvantages. Firstly, LiCoO_2 uses only half of its theoretical capacity, which arises from the fact that reversible lithium deintercalation/ intercalation occurs only in the $\text{Li}_1\text{CoO}_2 - \text{Li}_{0.5}\text{CoO}_2$ range, which gives low capacity of 130 mAh/g. Moreover, LiCoO_2 is environmentally unsafe and relatively expensive. Secondly, LiNiO_2 possesses higher reversible capacity (190 mAh/g) than LiCoO_2 , however, it is difficult to obtain ordered structure, due to strong cation mixing, which deteriorates transport and electrochemical properties. What is more, at high deintercalation level chemical stability worsens and exothermic reaction with liquid electrolyte takes place. Therefore, major investigation in this field focuses on solid electrolytes (mainly organic polymer type), which are to replace employed till now liquid one (EC/DMC- LiPF_6). Improved electrolyte/cathode interface would extend battery lifetime and improved operational safety. Thirdly, manganese spinel LiMn_2O_4 , used to be considered as a promising cathode material, suffers from decrease of capacity together with charge/discharge cycle number. This arise from its limited stability towards liquid electrolyte, leading to partial dissolution and decomposition coupled with disproportionation of manganese Mn^{3+} according to reaction (4):



Increase of chemical stability of manganese spinel may be achieved by proper modification of surface or change of bulk chemical composition - substitution of oxygen by fluorine or sulphur [18,19].

Complex research on oxide cathode materials for Li-ion batteries conducted by the author [6-8] points towards serious and diverse possibilities of improvement of electrochemical properties of $\text{Li}(\text{Co},\text{Ni},\text{Mn})\text{O}_2$ layered oxides, as well as enhancement of their chemical stability and reversible capacity. Those oxides show exceptional ionic-electronic transport properties, which are even improved in the process of lithium deintercalation, due to the occurrence of semiconductor-metal transition [6]. These results establish new criterion of effectiveness of the intercalation process and stability of mixed-valance systems. The obtained results allow to elaborate qualitative electron diagrams of such materials, explaining differences in electrochemical behaviour and chemical stability of cathode materials. Figure 7 shows qualitative model of electron structure of Li_xCoO_2 and Li_xNiO_2 cathode materials. Difference in overlap of 2p oxygen band with 3d cobalt (Figure 7a) and nickel (Figure 7b) band is shown.

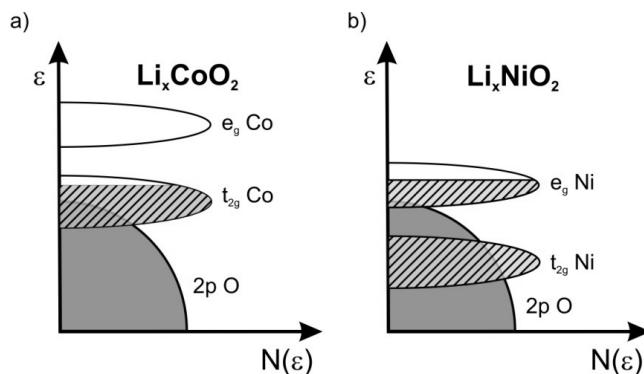


Fig. 7. Electron structure of (a) Li_xCoO_2 and (b) Li_xNiO_2 .

The observed difference in position of electron bands in both oxides (Figures 7a and 7b) is the reason of difference of their chemical stability upon electrochemical deintercalation. In the case of LiCoO_2 low spin configuration of Co^{3+} ions ($3d^6$) gives $t_{2g}^6 e_g^0$ orbital occupancy. This is responsible for low values of electrical conductivity and their activated character. Existence of electrical conductivity activation energy indicates presence of 0.7 eV energy gap between t_{2g} Co and e_g Co bands (Figure 7a). During electrochemical deintercalation of lithium, electrons (together with equivalent number of lithium ions) are taken from t_{2g} Co band. Electron holes are formed in t_{2g} Co band and the Fermi level is located inside it, leading to beneficial metal-like transport properties. Further deintercalation of lithium causes lowering of the Fermi level inside t_{2g} Co band, and when it reaches 2p O band, oxidation of oxygen ions takes place leading to oxygen release from Li_xCoO_2 structure. The crystal structure transforms from O3 to P3 type, which occurs for $x_{\text{Li}} = 0.5$ and reduces technological range of available capacities to half of its theoretical range. Released oxygen reacts with organic electrolyte (exothermic process) and threaten with battery explosion. Measurement of oxygen content and average cobalt oxidation state in samples at high deintercalation stage, as well as theoretical calculations by Ceder [20], confirm such model showing decrease of average oxidation number of oxygen with decrease of lithium content in Li_xCoO_2 . Spectroscopic measurements [21] confirm that electrons are taken rather from 2p oxygen band than 3d Co ($\text{Co}^{3+/4+}$) band.

In the case of LiNiO_2 , for which 2p oxygen band lays just beneath $e_g\text{Ni}$ band (Figure 7b), during electrochemical deintercalation of lithium electrons are removed from $e_g\text{Ni}$ ($\text{Ni}^{3+/4+}$) and then, in case of deep deintercalation $\Delta x_{\text{Li}} = 0.8 - 0.9$, electrons from 2p oxygen band are removed. This means that Li_xNiO_2 -based

material can work reversibly in far wider range of Δx_{Li} than Li_xCoO_2 , thus possessing higher reversible capacity.

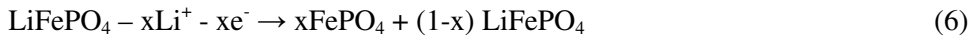
Wide range research carried out on optimisation of cathode material based on layered transition metal oxides in prof. Molenda's group [6-14] are directed towards the enhancement of chemical stability by introduction of manganese into cathode material ($\text{LiNi}_{1-y-z}\text{Co}_y\text{Mn}_z\text{O}_2$). Introduction of manganese affects material because of two factors: electron configuration $\text{Mn}^{3+/4+}$ ($t_{2g}^3 e_g^{1/0}$) and shift of 2p oxygen band out of the Fermi level variation region ($\text{Co}^{3+/4+}$, $\text{Ni}^{2+/4+}$) upon deintercalation/intercalation. At the same time, research are directed towards providing optimal ionic-electronic transport conditions, necessary for high effectiveness of intercalation.

There is continuous seek for cathode material based on iron compounds. This is because iron is inexpensive, abundant and is more environmentally benign than cobalt, nickel or manganese. However, simple LiFeO_2 oxide with layered structure (isostructural with LiCoO_2 or LiNiO_2) is unstable and has disadvantageously placed iron redox potential versus lithium [22]. $\text{Fe}^{3+}/\text{Fe}^{4+}$ redox potential is positioned too far from Li^0/Li^+ potential and exceeds electrolyte's electrochemical window, which makes it unstable towards electrolyte. At the same time $\text{Fe}^{2+}/\text{Fe}^{3+}$ potential is too close to Li^0/Li^+ potential, which leads to too low cell voltage. Such behaviour is owing to high spin configuration of Fe^{3+} ions and strong interactions between d electrons. Issues connected with instability of structure and disadvantageous position of redox couple of iron versus lithium can be avoided through transition to new series of polyanion iron compounds: $\text{LiFe}(\text{XO}_4)^+$ ($\text{X} = \text{S}, \text{P}, \text{As}, \text{Mo}, \text{W}, \text{y} = 2,3$) [22], with large polyanions stabilizing the structure.

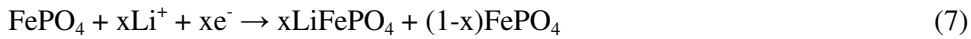
Large hopes are placed on LiFePO_4 with olivine structure (Figure 5), commonly known as phospho-olivine, which possesses significant advantages: high theoretical capacity (170 mAh g^{-1}), the highest chemical stability among all known cathode materials – guaranteeing operational safety, and lack of deterioration of capacity with number of charge/discharge cycles. In its crystal structure dense hexagonal close packed oxygen sublattice forms one-dimensional potential fast lithium ion diffusion paths. However, structural conditions (corner-shared FeO_6 octahedra give substantial Fe-Fe distance, around 4\AA , whereas conducting oxides has M-M distance below 3\AA) cause that this material is practically electron insulator. Its electrical conductivity at room temperature, i.e. at operational temperature, is extremely low as for a cathode material, and reaches $10^{-9} \text{ S cm}^{-1}$ (a few orders of magnitude lower than that of LiCoO_2). These features cause that observed mechanism of LiFePO_4 delithiation is not a diffusional process, which would be according to reaction (5) and lead to homogeneous material with variable lithium content:



Available literature data, as well as our own research, however, indicates that lithium extraction from LiFePO_4 during charge process is decomposition of cathode material into two phases: containing lithium and without it:



and discharge:



Reversibility of charge and discharge processes is connected with close similarity of LiFePO_4 and FePO_4 crystal structures – they both are described with *Pnma* space group and unit cell volume differs only 6.81%. Such mechanism of delithiation is strongly disadvantageous – only grain surface works effectively, which determines low current density. Low ionic-electronic conductivity is a major reason of two-phase mechanism of delithiation.

High hopes for lithium batteries market arose after tremendous reports by Chiang et al. concerning doping of phospho-olivine [23], which indicated possibility of increase of electrical conductivity as high as 10^7 times. Also theoretical works appeared, showing that metallic-like properties are possible in phosphoolivine. At the same time surface studies of doped phospho-olivines carried out in J. Molenda's group [24], as well in L.F. Nazar's group [25] showed that high conductivity of doped phospho-olivine is not a bulk feature, but rather are due to iron phosphides precipitated at surface in the process of partial reduction of LiFePO_4 to Fe_2P during synthesis. This does not improve current efficiency, since it does not provide bulk ionic-electronic transport, necessary in diffusional deintercalation/intercalation of lithium (Eq. 5).

The main issue in research on cathode material based on phospho-olivine is to obtain phospho-olivine with mixed ionic-electronic conductivity, which should commence diffusional mechanism of deintercalation / intercalation and improve current efficiency lithium cells based on phospho-olivine, which now is the only drawback of such batteries. Improvement of electrical conductivity of LiFePO_4 is the main goal of LiFePO_4 -based cathode material's technological research.

Intensive research in prof. Molenda's group on improvement of transport properties of phospho-olivine i.e. on obtaining phospho-olivine with mixed ionic-electronic transport [26] shows that this can be achieved by substitution of iron with transition metal with suitable electron configuration in respect to iron, in order to generate electron transfer from one ion to another. It seems that further possibilities of improvement of ionic-electronic transport exist not only

by substitution of iron, but also by modification of polyanion $(\text{PO}_4)^{3-}$. Another way of improvement is reduction of particle size and going down to nanoscale, which could shorten effective lithium diffusion distance and improves current efficiency of a cell. Materials engineering methods allow to produce this material with highly electrochemically active nanograins with uncovered lithium fast diffusion paths.

Nowadays, phospho-olivine is in the centre of interest of large companies producing lithium batteries, which are already manufacturing Li-ion batteries based on LiFePO_4 (A 123, Saft, Valence,, Samsung, Sony and others). China starts mass production of LiFePO_4 , having in mind that it is 10-times less expensive than currently employed cathode materials based on LiCoO_2 . Such policy can give them control over global Li-ion batteries market.

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CURRICULUM VITAE



Janina Molenda, Prof. Ph.D., D.Sc., Graduated from AGH University of Science and Technology, in the field of Materials Science in 1975, received her Ph.D. from AGH University of Science and Technology in 1979 and from Université Bordeaux I, Bordeaux, France in 1984. Obtained habilitation from AGH University of Science and Technology in 1988, where currently holds the position of Head of Department of Hydrogen Energy at Faculty of Energy and Fuels. Prof. Molenda is also President of Polish Hydrogen and Fuel Cells Association. Her main fields of interest are Solid Oxide Fuel Cells (transport properties of cathode materials, catalytic properties of perovskites, defect structure and ionic conductivity of ceria based electrolytes), lithium-ion batteries (cathode materials - LiFePO_4 , doped

LiMn_2O_4 , LiCoO_2 based materials, anode materials), nonstoichiometric compounds, fundamental studies of transport properties of solids, metal-insulator transitions, HT_c superconductors, electrochromic effect, fundamental studies of intercalated cathode materials ($\text{Li}_x\text{M}_a\text{X}_b$, M - transition metal; X = O, S, Se) in order to correlate structure and electronic properties with effectiveness of intercalation process.