

Review

Battery and/or supercapacitor?—On the merger of two electrochemical storage system families

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Abstract: Similarities and analogies between materials, structures, operating and construction principles of secondary batteries and supercapacitors and their electrodes are presented, named, and reviewed in context. On the material level, several materials used both in batteries and supercapacitors are addressed, and implications from observations made in one application for the other are highlighted. On the electrode level, a continuous change of architectural details is observed when going from an electrode with high charge storage capability to an electrode supporting high currents is detected; again, this overlap provides instructive ideas for both fields. On the cell level, combinations of electrodes from both fields yielding hybrid devices are an obvious outcome again, with implications for both fields. Ideas and suggestions for further research and development based on a deeper exchange between both families are developed.

Keywords: battery electrodes; supercapacitor electrodes; capacitive behavior; pseudocapacitive behavior; electrochemical energy storage; electrochemical energy conversion

1. Introduction

Electrical energy can be stored by various means and procedures [1]. A fundamental difference between the various principles and systems of electrical energy storage (EES) is related to the question of whether storage proceeds directly without any conversion process or whether such a mechanical, physical, or chemical conversion process of, e.g., electrical energy into chemical energy is required. The essentials are schematically depicted in **Figure 1**.

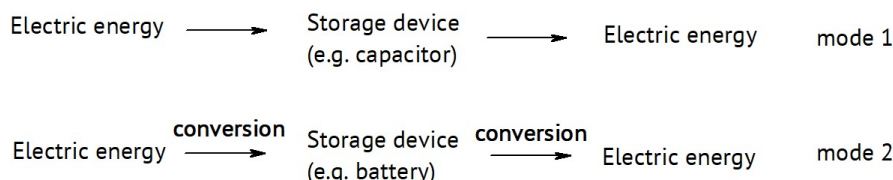


Figure 1. Basic modes of EES.

A typical, prominent, and highly successful example of the first mode is the capacitor; an equally popular and successful example of the second mode is the rechargeable battery. Discoveries of fundamental principles like static electricity for the former device as well as inventions like the lead-acid battery for the latter one are items in the history of science and technology frequently retold in textbooks and

reviews [1,2]. The very different origins of these two principles kept both technologies apart from each other, with only minor exceptions of overlaps or interactions like the electrochemical treatment of aluminum foils for electrolytic capacitors. Although the latter device has even more connections to electrochemistry than a plain surface treatment, such capacitors have never been considered electrochemical energy storage devices for a simple reason: Storage in capacitors without any conversion reaction is based simply on the separation of electric charges on two metal plates (electrodes), whereas storage in a battery is based on a faradaic (redox) reaction with associated more or less extended chemical transformations of the electrode materials (for a scheme and an example, see **Figure 2**).

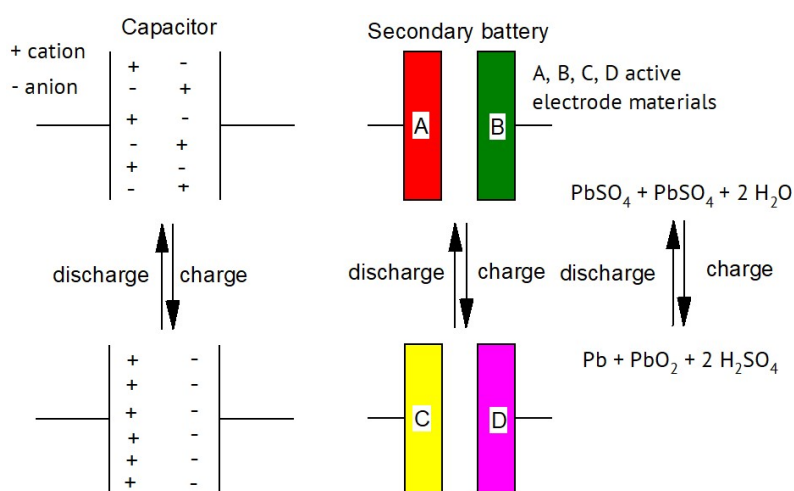


Figure 2. Schematic principles of operation of a capacitor and a secondary battery (with a typical example).

The electrochemical double layer (EDL) established at the interface between an electronic conductor (e.g., a metal electrode) and an ionically conducting phase (an electrolyte solution) is not a recent observation but has been studied, modeled, and measured for decades [3]. Finally, the reports and patents by Becker [4] and Rightmire [5] have moved this omnipresent capacitive property of an electrochemical phase boundary into the view of electrical engineers. This development resulted in the now very popular supercapacitor¹ of the electrochemical double layer capacitor-type (EDLC) [6]. Although the storage principle is again the separation of electric charges, like with the conventional capacitor, the participation of ions in the electrolyte solutions balancing the electric charges in the electrode (the electronically or the hole-conducting conducting phase) has made supercapacitors a highly attractive research topic for electrochemists, although no faradaic or redox reactions proceed at the electrode during storage and release of electrical energy.

Supercapacitors have been established as means of EES in a vast range of sizes, from tiny ones in environmental and medical applications across numerous mobile applications, e.g., telecommunication, to large ones in electric vehicles and supply systems in mass transit. Introductory overviews are available [7–13], in numerous monographs, presumably all aspects, from basic functional principles on to materials and applications, are treated [14–22]. A review of self-discharge as a major drawback of EDLC devices is available [23]. The essential role of secondary batteries is stressed

almost everywhere in the introduction of every publication on topics related to them; their still-growing importance in mobility and in the use of renewable energies is well-established [1,2,24].

Because the storage capability of an EDLC is fundamentally limited by the size of the interfacial area (between the electronically conducting phase, the electrode, and the ionically conducting phase [25]), the amount of charge possibly stored in such a device is limited, and accordingly, the energy, too.

The observation that some surface-confined redox reactions on electrode surfaces show a current response to a changing electrode potential very similar to that of a capacitor (they behave almost like the electrochemical double layer) [26] has stimulated still-booming research into redox-active materials for supercapacitor electrodes. Some of these materials are also encountered in primary and secondary batteries; the design principles as well as electrode chemistries of some battery and supercapacitor electrodes are very similar or just the same. This fact has initiated a discussion about the possibly ongoing merger of secondary batteries and supercapacitors. Or, in other words, secondary batteries and supercapacitors are basically based on the same principles, differing only in details. The overlap and/or merger may proceed if this observation is correct beyond the author's personal opinion on various levels beyond those already mentioned: Combination of battery electrodes and supercapacitor electrodes in advanced lead acid systems, as suggested by Shukla et al. [27], on the device or cell level, as well as use of electrodes containing both a redox-active electrode material and a capacitive one [1] on the electrode level. These concepts have been sometimes named hybrid ones for both levels [28,29]. On the cell level, the term asymmetric frequently shows up [30]. The term "hybridisation" or "hybrid" appeared in the present context some time ago [31]. Some of the resulting electrodes and full devices mostly discussed in terms of hybridisation may actually be called the results of said merger, but the term has not been applied before. Given the absence of a clear definition of this term, it does not surprise that this report deals mostly with combinations of battery and supercapacitor electrodes (called asymmetric devices elsewhere); for an example, see below. Some further thoughts related to distinguishing between supercapacitors and batteries have been provided elsewhere [32].

Beyond the conceivable merger of two fields and their constituents, some confusion regarding terminology and understanding seems to be looming, too. Generous creation of acronyms adds to this. The temptation to create further terms in a sometimes rather vain attempt to create something quotable has resulted, among others, in positrode and negatrode (apparently instead of positive and negative electrodes, simply) or supercapattery naming a device combining aspects of secondary batteries and supercapacitor behaving more like a supercapacitor (i.e., have somewhat higher power output) and supercabattery (the same, but somewhat higher energy output) [33–35].

Whereas in „electrochemical double layer capacitors“ the principle of the electrochemical double layer, in particular its capacitive behavior, is utilized (see **Figure 2**) in supercapacitors of the redox type and their materials discussed in this report, redox processes, in particular those proceeding close to the electrode/electrolyte (solution) interface (so-called superficial, see **Figure 3**), are used

in addition for charge and subsequently energy storage [36,37]. In contrast to battery electrodes, wherein redox transformations proceed all over the volume (at least in the desired ideal case of complete mass utilization), only the topmost layer(s) of the material is used for energy storage in a supercapacitor application. Nevertheless, an increase in storage capability by 2 to 3 orders of magnitude has already been estimated in the first reports suggesting this approach [37]. Typical advantages of the EDLC, in particular the extremely fast charge and energy storage and release because of the purely capacitive processes only limited in terms of current by the internal resistance (electrical series resistance (ESR)) and not impeded by any chemical or electrochemical reaction, are only slightly diminished when utilizing surface-confined processes in redox supercapacitors. As an alternative name, the term Faradaic supercapacitor [6] has been used².

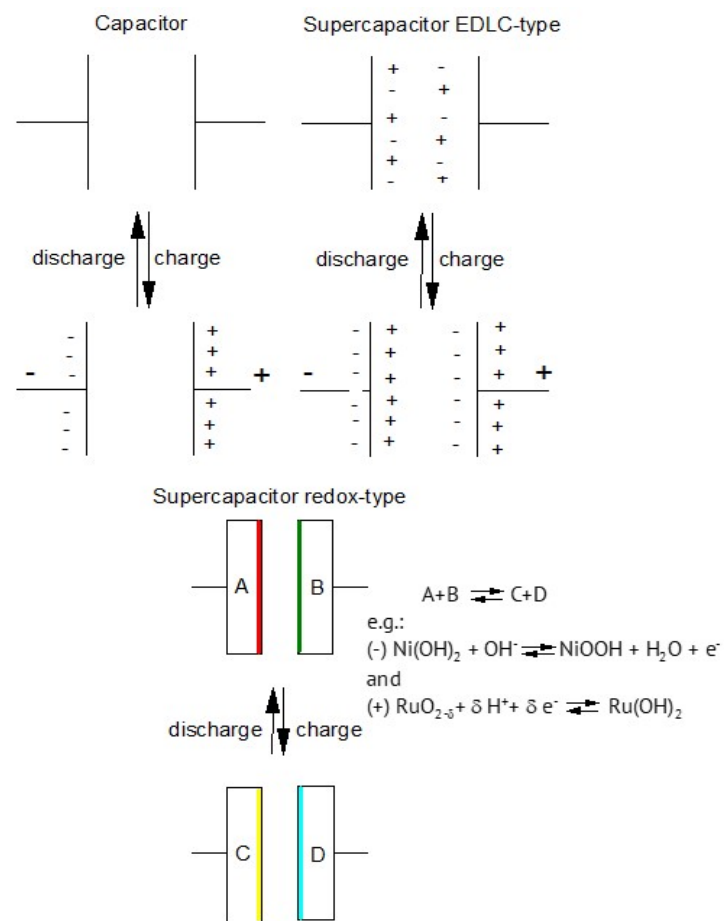


Figure 3. Charge distribution during operation in a conventional capacitor and in EDLC and redox-supercapacitors.

Reports on similarities and connections between the two families of energy storage systems seem to coincide with communications on differences and boundaries. Unfortunately, imprecise use of terminology sometimes ending in simple contradictions within a report has turned out to be more confusing than helpful [38]. A recent update has attempted to provide some clarification [39].

Following, we will inspect approaches, mergers, and overlaps between the two fields on various levels, starting at an atomistic or microscopic one and ending at the

device level. For the sake of clarity in this report, in a roughly simplified way, the term supercapacitor (SC) indicates an EES device providing high current (rate) capability, whereas the term secondary battery names a device with high energy storage capability. Further overlaps between aspects of both systems, like manufacturing processes, were addressed elsewhere [40].

Starting from the implicit hypothesis that redox supercapacitors and secondary batteries are actually only modified versions of the same device, this report aims at highlighting common and separating aspects of both fields, and it shall help in particular to enable “crosspollination” between the fields by suggesting possible transfers of ideas, methods, and concepts from one field into the other.

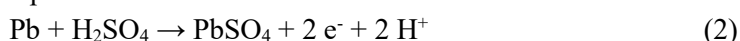
2. Chemistries, reactions, and processes—merger at the atom and molecule level

Storage of electrical energy can proceed in the simplest case of the electrostatic capacitor by separating charges. The work (energy) spent to achieve this is released again upon discharge when the temporarily separated charges are redistributed again (see **Figure 2**). This applies to the traditional capacitor in all its commercial types; it also applies to the electrochemical double-layer capacitor. The only distinction between both is the fact that in the conventional capacitor, only one charge separation across the device proceeds, whereas in the EDLC, charge separation proceeds at both electrodes (see **Figure 3**).

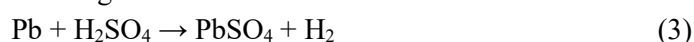
In a battery, energy is stored by using electric energy to drive a chemical transformation. The obtained materials are “richer in energy” (the absolute value of the Gibbs energy or free reaction enthalpy is larger) than the starting materials. As an example, the charging reaction at the negative electrode of a lead-acid battery shall be considered:



Upon discharge the process is reversed:



As an alternative the following reaction is conceivable



Assigning an energy or enthalpy to both reactions (Equations (1) and (2)) is thermodynamically correct. The Gibbs energy (free reaction enthalpy) of the first process can be calculated, yielding $\Delta G_{\text{R}} = 68.7 \text{ kJ}\cdot\text{Mol}^{-1}$ for the charging reaction. For the discharge reaction, $\Delta G_{\text{R}} = -68.7 \text{ kJ}\cdot\text{Mol}^{-1}$ is obtained. In reality, the first process will not occur without applying external force. Equation (3) describes an unwelcome corrosion reaction with the two electrons used to reduce protons, but this is not the purpose of an electrode in a rechargeable battery. Quite obviously, one electrode (half-cell) does not make up a full cell; a second electrode is needed. This very simple consideration makes all considerations and calculations frequently found in scientific reports about the energy assigned to a single electrode and its reaction irrelevant. Nevertheless, the authors feel some certainty based (if they have considered this at all) on the relationship between the free enthalpy and the cell voltage U_0 :

$$\Delta G_{\text{R}} = -n \cdot F \cdot U_0 \quad (4)$$

Although sometimes cell voltage U_0 (also called electromotive force emf) and electrode potential E are confused (some authors simply use one symbol for both), this equation certainly does not apply to an electrode. To escape this conundrum, an apparent solution is found by taking two electrode potentials from a charge/discharge experiment or a cyclic voltammogram (CV), calculating the electrode potential difference, and assuming this to be a cell voltage. But again, one electrode does not make up a cell. For lists of electrode potentials calculated from free enthalpies, it is always assumed that the second half-cell is the standard hydrogen electrode (for example, in the study by Wu and Holze [1], listings with references are available in the study by Holze [41]). At this point, it appears to be reasonable and highly recommended to assign only charge storage capabilities to a single electrode (material), possibly combined with the electrode potential, wherein this happens but no energy.

Chemistries and fundamental operating principles of both families met when first reports about the capacitive-like current response in CVs of some metal electrodes appeared and the specific current response towards a changing electrode potential was observed in CVs [37,42–45]. As an example to illustrate the phenomenon and concept (although without practical application value), CVs of a polycrystalline platinum sheet electrode in contact with an aqueous electrolyte solution of 0.05 M H_2SO_4 recorded at different scan rates are shown in **Figure 4**.

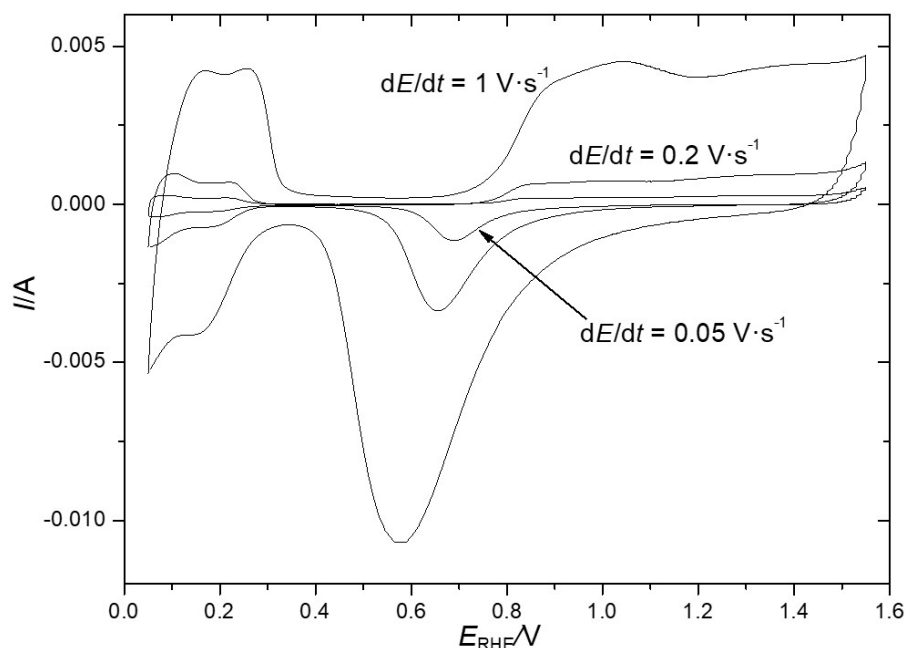


Figure 4. Selected CVs of a polycrystalline platinum electrode in contact with an aqueous electrolyte solution of 0.05 M H_2SO_4 , nitrogen purged, scan rates as indicated.

Evaluation of the current response in the double layer region where no Faradaic process is observed (i.e., $0.4 < E_{\text{RHE}} < 0.6$ V) and of the current in the electrode potential region where formation of a hydroxide/oxide coverage proceeds (i.e., $0.9 < E_{\text{RHE}} < 1.5$ V) yields the following **Figure 5**.

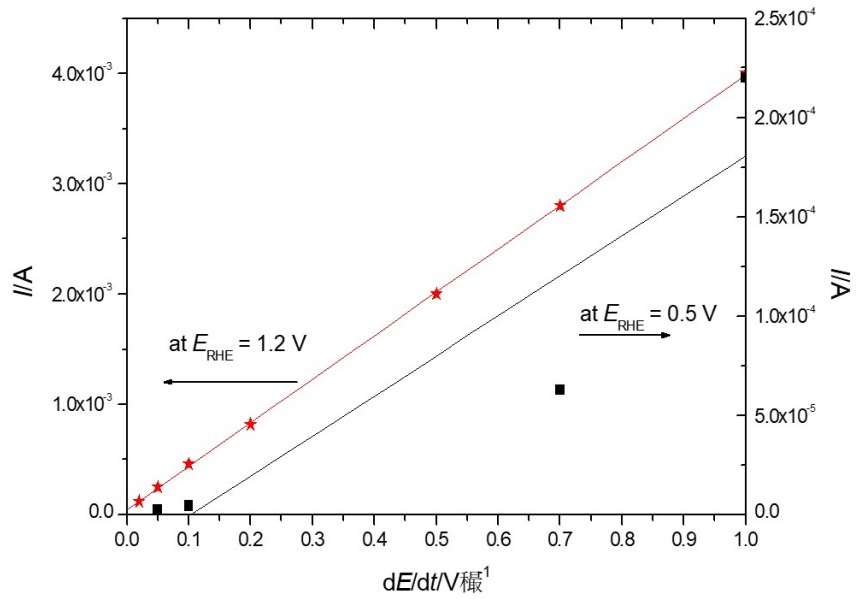


Figure 5. Current response vs. scan rate of a polycrystalline platinum electrode in contact with an aqueous electrolyte solution of 0.05 M H₂SO₄, at different electrode potentials.

Data at both electrode potentials is taken from the same cycle. Values recorded at $E_{\text{RHE}} = 0.5 \text{ V}$ are imprecise in this approach; much more precise data found in a narrow range of electrode potentials with potential scans limited to the double layer region only have been reported before [7,12,46].

From the slope of both lines (dI/dv), a differential capacitance C_{diff} can be obtained. In the case of values noted at $E_{\text{RHE}} = 0.5 \text{ V}$, this will indeed be the double layer capacitance C_{DL} , which in turn can be used as a measure of the electrochemically active surface area (EASA) [46,47]. The currents found at $E_{\text{RHE}} = 1.2 \text{ V}$ are bigger by more than an order of magnitude and cannot be explained this way. They are due to the formation of hydroxide/oxide coverage, although the current response looks like that of a capacitor. The term *pseudocapacitive* presumably coined by Gileadi and Conway [42], simply and generally describes a response of the electrode like that of a capacitor, as shown in the preceding figures, as already suggested by the term (Greek: ψεύδειν (Pseudes): pretending, fake; in the present content, the meaning is as used in biology and botany for designing a species with the name pseudoxxx because it looks like xxx is obviously intended). The relationship between scan rate and current response can be stated as

$$I = C_{\text{diff}} \cdot dE/dt \quad (5)$$

or using $v = dE/dt$

$$I = C_{\text{diff}} v \quad (6)$$

This can be taken quite obviously as a specific case of a general power law relationship

$$I = a \cdot v^b \quad (7)$$

with $b = 1$ being typical of a capacitive process and behavior. In the case of the current response in the double layer region with $b = 1$ and $a = C_{\text{diff}}$, at $E_{\text{RHE}} = 1.2 \text{ V}$, the response is due to a redox process, whereas the response is still behaving like a capacitive one, thus the suggested designation. At this point, it can be concluded that

the term *pseudocapacitive* is purely descriptive (and certainly makes sense only as an adjective). The extensive discussion published in numerous reports on extended terminologies of pseudocapacitance (s) appears to be rather unproductive because it only creates further terminology but fails to provide an understanding of the reasons for the current response with some materials and processes, but not all redox processes at electrodes. Attempts to get an understanding of the causes have been discussed before [26]. Meaning as well as proper application of the term “pseudocapacitive” have been subjects of heated debates [26,38,48–51]. A previous use of the term “pseudocapacity” by Grahame [52] means redox processes at the mercury electrode/aqueous electrolyte solution interface with reduction and subsequent oxidation of cadmium/cadmium ions; thus, this meaning handles solution species differently from the suggestion by Gileadi and Conway involving redox reactions of surface-attached species.

Various processes may constitute the Faradaic reaction causing this response beyond this example:

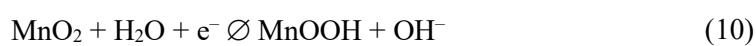
A simple redox process involving species on the electrode surface involving the electrode material itself:



Or a redox process of an electrochemically active coating [26].



Or a redox reaction of an electrode material:



In all cases, material and system, i.e., electrode, properties discussed elsewhere are responsible for the capacity-like (*pseudocapacitive*) current response and the absence of current peaks frequently observed in CVs obtained with redox active materials, as discussed in detail before [26]; for further considerations, see, e.g., Chen [55]. The last example (Equation (10)) already shows an overlap with battery electrode materials: Manganese dioxide is a frequently encountered material in many, mostly primary, systems [56,57]. Some materials like $\text{RuO}_2 \cdot 0.5\text{H}_2\text{O}$ show this pseudocapacitive behavior always, i.e., independent of particular preparation, morphology, particle size, etc.; they have been called intrinsically pseudocapacitive [58], whereas materials showing this property only after some “engineering” (e.g., nanostructuring, composite formation) have been called extrinsically pseudocapacitive.

All examples share common features also encountered in battery electrode chemistries and processes:

The participating species must be insoluble in the electrolyte solution.

The processes should be fast, in the frequently assumed meaning of reversible electrode kinetics as being fast.

The involved species should be readily available; even better, they should be abundant.

Preferably, they should be cheap, non-toxic, and environmentally compatible.

These general requests are already pointing into the next section, where they will be extended and specified, but some details more related to processes require attention here: Following the first patent [59], the use of redox-active, soluble species added to

the electrolyte solution was recommended in particular for supercapacitors of the EDLC-type, apparently in contradiction to request 1. As pointed out before, this setup is basically a redox flow battery without the flow [60], and without a separating membrane or an equivalent device, keeping the electrolyte solutions operating at the positive and negative electrodes from mixing may be very remarkable [23]. With a membrane instead of the highly porous separator, the internal resistance will grow, making such a device rather unattractive, at least for high-power applications. In terms of operating principle, it brings the redox flow battery and thus the idea of a battery with soluble electrode materials (e.g., the lead acid battery with soluble active materials [1,61]) into the picture. Thus, this concept is one more example of the ongoing merger. The initial concerns regarding fast self-discharge, in particular of an undivided device, can be put into perspective when considering the application: A supercapacitor used only for very short-term energy storage may not have its practical value significantly diminished by self-discharge.

Beyond the two principles of (1) charge separation in a capacitor of whatever kind and (2) a redox reaction, hardly any other process appears to be suitable to be involved in charge and thus energy storage. Nevertheless, new terms like “intercalation pseudocapacitance” were created (for an example, see the report by Lou et al. [62]). Whether they really help is doubtful and should not be discussed here; their inventors seem to overlook that “pseudocapacitive” simply describes a particular current response to a changing potential or voltage without asking for specifics of the underlying electrochemical and possibly associated further chemical processes. Because intercalation is indeed a term encountered when examining battery electrode materials, the common options of processes associated with an electrochemical charge transfer in battery electrodes should be inspected:

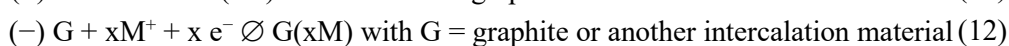
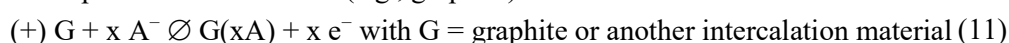
- Intercalation;
- Alloying;
- Conversion.

Intercalation (the term insertion is sometimes also used with intercalation, mostly applied to layered materials) itself is not an electrochemical process but a chemical reaction. Coupled with the ingress or egress of ions like, e.g., Li^+ in the negative (graphite) and positive (metal oxide) electrodes of a lithium ion battery, it is a frequently encountered material storage option currently operating in many electrodes of lithium ion batteries and capacitors. Because structural changes in the host material are mostly small, the process may be fast and thus compatible with the fast electrode reaction required for a supercapacitor. Among the many processes and associated materials (in particular metal oxides) suggested for supercapacitor electrodes, verified examples of intercalation were not reported. Actually, the electrode reactions are not even known exactly in most cases [63]. Graphite electrodes used for hosting lithium ions as employed in lithium-ion batteries are used as negative electrodes in lithium-ion capacitors; because they show no pseudocapacitive behavior, they are not considered here.

Alloying is a storage option for, e.g., silicon or tin electrodes in lithium-ion batteries. Because of the extensive structural changes, this storage process appears to be hardly suitable for supercapacitors. The reported current responses in CVs certainly do not suggest pseudocapacitive behavior.

Conversion processes as encountered with, e.g., the lead electrode in a lead-acid battery show high material utilization, and the processes inspected above for supercapacitor electrodes can be taken as typical examples despite the fact that the proceeding reactions will cause major structural changes when happening in the bulk of the electrode material. Such changes in a film of hydrous $\text{Ru}(\text{OH})_2$ or a thin coating of electrodeposited MnO_2 are obviously no significant problem given the reported stability of related supercapacitor electrode materials.

There is a family of batteries commonly called dual-ion batteries³ with some subspecies [64–70] wherein the redox transformation associated with charge storage as well as other conceivable modes of charge storage are not as obvious as indicated in Equations (1) and (2). In its most basic form, it is an ion intercalation/deintercalation battery with the cations M^+ and anions A^- of the electrolyte (MA) going in and out of the respective host materials (e.g., graphite).



Instead of intercalation/deintercalation adsorption/desorption with the associated advantage of a high rate is conceivable [70], but a possible pseudocapacitive behavior moving such cells into the focus of this report has not been noticed.

The merger of battery and supercapacitor chemistries has been reviewed elsewhere [28].

3. Materials

Processes and operating principles outlined in the preceding section need materials enabling them; in addition, these actual materials are subject to further consideration. Electrode materials for both battery and supercapacitor electrodes (in the latter case of the redox-type) should meet certain well-established requirements, some of them more related to processes and chemistries than were already addressed in the preceding section:

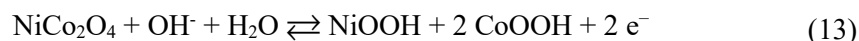
For practical applications, various properties of the active masses are relevant and desirable:

- Cheap and abundant raw materials;
- Environmental compatibility;
- Fast and chemically as well as electrochemically reversible electrode kinetics;
- Mechanically stable structural integrity during charge/discharge;
- Chemically as well as electrochemically stable under all operating conditions (temperature, current);
- Sufficient electronic conductivity
- Morphology: providing a large interfacial area with electrolyte solution;
- Optimized porosity enables fast ion movement;
- Compatible with electrolyte solutions and other electrode and cell components.

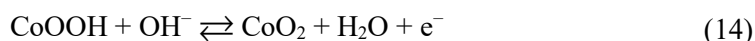
3.1. Chalcogenides

This “wish list” has been common in battery research for decades; it applies as well to redox supercapacitors. Currently, predominantly compounds from the metal chalcogenide class of materials with a single metal (e.g., MnO_2), two or even three

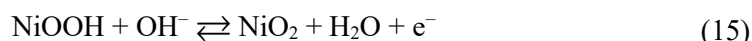
metals (binary chalcogenides, like $\text{Me}_1\text{Me}_2\text{O}_z$ and related oxides like $\text{MeMe}_1\text{Me}_2\text{O}_z$ with Me1 and Me2 most frequently being transition metals, e.g., CoFe_2O_4) are encountered. With materials containing more than one kind of metal, the question of the proceeding redox reactions might be asked. Given the vast number of possible combinations of two or even three different metals, many possible redox processes, their relative likelihood, their reversibility, and their contribution to overall charge storage cannot be discussed here; this will be reported elsewhere [63]. As an example, NiCo_2O_4 studied by Wang et al. [71] shall be considered. The authors proposed the following redox reaction:



which hardly can be correct; a simple calculation shows that only one electron is released. It appears also rather unlikely that nickel cobaltite with a specific crystal structure disintegrates into two metaloxy hydroxide compounds and reassembles into the cobaltite again on the way back. In the synthesis of the material, high-temperature calcination is required for this to occur. As a follow-up reaction, Equation (14) was proposed.



No reason was provided why the reaction in Equation (15) should not proceed.



Taking this reaction into account, four electrons may be transferred when all metal ions change from their initial state of oxidation into their highest conceivable state of oxidation. It is rather astonishing to notice that there is no report addressing this question in detail, preferably using *in situ* methods sensitive to the state of oxidation. Even *ex situ* studies with, e.g., XPS possibly providing evidence of the state of oxidation of the participating metal ions at various electrode potentials where the materials were emersed from the electrolyte solution are not available.

The use of redox-active materials for charge storage instead of activated carbon or other carbonaceous materials for double-layer storage to increase storage capability comes at a price: Decreased current capability and lower stability of the materials. The former is mostly due to lower electronic conductivity and thus increased internal ohmic resistance of the material, as well as the limited rate of the redox reaction. This aspect depends on the actual material; the actual effect can be ameliorated to some degree by, e.g., nanostructuring. The reduced stability is mostly due to volume changes of the active material because redox reactions are frequently associated with further chemical and structural changes (see above, Equation (1), etc., and the report by Dubal et al. [72]). Again, suitable structuring of the active material and/or combination with auxiliary materials may help both battery and supercapacitor electrodes. A particularly popular combination includes a redox active material and some form of carbon prepared as a composite with, e.g., MnO_2 deposited on a carbon support of suitable porosity (for an overview, see Borenstein et al. [73]). Such a combination, going beyond the conventional mixing or blending of powders of active mass with, e.g., acetylene black, may even be called a hybrid. Although it appears that the term “hybrid” enjoys a rather widespread and poorly defined usage, it seems safe to state that such deposition of a chalcogenide on a mesoporous carbon support may result in

a product with properties and performance going beyond a simple addition. Whether it is justified to discuss such interactions presumably causing these effects in terms of interfacial conjugation [55] seems to be another question.

3.2. Intrinsically conducting polymers

A second class of materials considered both for use in battery and supercapacitor electrodes encloses intrinsically conducting polymers (ICPs) [74–78]. Starting with the suggestion of the charge storage possibilities of ICPs possibly useful in electrodes for secondary batteries (which unfortunately was not crowned with a commercial success until today), these materials have also been suggested as electrode materials for supercapacitors [78]. Chances and challenges regarding the properties and performance of the materials are the same in both applications, and ways to establish the desired long-term stability during multiple charge and discharge processes are even more mandatory in the case of supercapacitor applications. Accordingly, in the latter case, the architecture of the ICP and suitable morphology, possibly improved by combination with a matching second component, deserve increased attention. Suggestions that ICPs will fill the gap between batteries and supercapacitors may be a bit too optimistic [78].

A class of materials attracting growing attention for both battery and supercapacitor electrodes are organic materials beyond ICPs. The recent surge in interest is presumably related to the general interest in materials with practically unlimited resources (which is a growing concern with many current battery materials), to the possibility of rather simple handling of used/worn out materials not requiring the specific procedures required for handling heavy metal-containing batteries and devices, to the simple redox reactions not encumbered by intercalation or other possibly slow reaction steps, and to mostly smaller energy usage in preparing these materials under environmentally acceptable conditions (for a broader overview with regard to metal-ion batteries see the overview by Lu et al. [79], for some rather general considerations on organic materials in batteries see the report by Huang et al. [80]). Changes in material properties can be afforded in most cases using the tools of well-established organic synthesis [81]. Finally, these materials may provide a bridge to the use of renewable materials like lignin [82] or other materials derived from natural resources [88–90]. Lastly, these materials may enable or at least simplify the construction of flexible and even stretchable battery and supercapacitor devices.

3.3. Composites and further combinations

As already discussed above, for the case of combinations of chalcogenides and carbon materials, combinations of ICPs and some carbon-based components were examined. Several reviews are available [91–96]. Although not always addressed specifically, there are several roles for both the ICP and the carbon component. The ICP will predominantly act as the charge storage material. As a polymer, it might also act as a binder, caring for both cohesion between the ICP in its particular shape and the carbon in the electrode mass and adhesion of the composite or hybrid to the current collector. The carbon provides a mechanical support in case the ICP has been deposited on it; it provides mechanical integrity when the ICP shows shape change

because of, e.g., swelling and shrinking; and it guarantees sufficient electronic conductivity even when the ICP is in its poorly conducting neutral state.

Finally, combinations of metal chalcogenides and ICPs have attracted attention as active masses, mostly for supercapacitors; a review has been provided by Fu et al. [97].

Supercapacitors as well as battery electrodes frequently contain auxiliary materials. In addition to the current collector almost always also acting as a mechanical support (metal foils, carbon, or graphite paper), a binder and a conductive additive (acetylene black or other carbonaceous materials) are present. In both applications, materials are needed: without a binder, coherence between the particles of the active mass as well as their adherence to the current collector would be insufficient for keeping the electrode in shape and stable during cycling. Without added conducting material, the internal ohmic resistance (sometimes confused with the charge transfer resistance R_{ct} or assumed to be almost equivalent to it) of the active mass would be too high to support the high current operation requested both for supercapacitor and high-power battery electrodes. To get rid of the latter additive, mixed metal chalcogenides with higher intrinsic electronic conductivity are attractive when compared with, e.g., the wide bandgap semiconductor forms of MnO_2 . In many reported studies, this potential advantage is hardly explored; instead, the almost standard additions of acetylene black, mostly around 10 %wt. indicate that no optimization was tried.

To get rid of the conventionally electrochemically dead binder, electrode preparation procedures wherein the active mass is directly deposited on the current collector (e.g., deposition of metal oxides and hydroxides by chemical or electrochemical procedures from the respective ionic solutions on metal foils, as suggested by the Lokhande group [98]) may provide an option at least for low loadings of active mass. These were identified as being particularly favorable for high mass utilization [99]. Electrochemically active binders based on ICPs have been suggested by Kondratiev and Holze [100]. A review by Holze and Kondratiev is available [77]. Another option is a polystyrene-based binder with redox-active modifications [101]. The possibility of using an ICP both as a redox-active storage material and as a binder when combined with a metal chalcogenide has hardly been explored so far because, in the majority of reported studies (a review by Fu et al. can be found [97]), the ICP has been polymerized chemically in the presence of the finely dispersed chalcogenide, yielding a powder material. To transfer this material into an electrode, a binder is needed. A procedure wherein electropolymerization in the presence of chalcogenide is performed appears to be feasible; it was examined before with promising results for nanocomposites in corrosion protection coatings [102].

4. Electrodes and their architectures

Based on the current response to a changing electrode potential in a CV experiment (or the electrode potential change during a galvanostatic charge/discharge (GCD) experiment), two types of electrodes can be straightforwardly distinguished:

Battery-type electrode (also called redox electrode or Nernstian electrode [103]): The CV shows visible current (Faradaic) peaks associated with the redox transformations of the active material; the GCD curve shows steps or plateaus.

Because electrodes with very few notable exceptions (lithium metal electrodes) are highly porous with an associated large surface area and interfacial capacitance, they will always have significant capacitive currents added to the Faradaic current, yielding CVs that look different from the textbook example of a CV obtained with a compact and smooth metal sheet in an electrolyte solution with a dissolved redox system.

Capacitive electrode⁴: The CV is flat; the GCD curve shows a triangular shape. Only ion accumulation and dispersion (possibly including ad-/desorption) proceed during charging and discharging (see **Figure 2**). Whether adsorptive interaction between the electrode surface and ions close to it happens or not may be of academic interest. Details of partial charge transfer and electrosorption valency are discussed [104]. A flat CV and a triangular GCD curve may also be obtained with an electrode showing pseudocapacitive behavior (see **Figure 4**). But the currents are larger by orders of magnitude than those found with comparable (in terms of EASA and dimension) simply capacitive electrodes; the larger currents cannot be due only to ion accumulation and dispersion but are due to mostly superficial redox processes that lack the peak in a CV (and the plateau in the GCD) for reasons related to materials and species properties discussed elsewhere [26]. Basically, again, a combination of Faradaic and non-Faradaic processes, like in # 1, happens, but this time the current vs. potential relationship is utterly different from the Nernstian case. This electrode may also be called a pseudocapacitive electrode.

Sometimes current responses, more specifically the shape of the recorded curves and diagrams, appearing to be somewhere between types 1 and 2, have been generously called capacitive or pseudocapacitive; this only causes confusion and does not help in understanding the underlying phenomena. Details of such curves have been amply displayed and extensively deplored elsewhere [105] without providing a deeper understanding of the underlying phenomena. Because of the great interest in having electrodes with both high charge storage and current-generating capability as outlined above and to understand relationships between structure, morphology, architecture, and performance, there have been frequent attempts to separate the Faradaic and the capacitive (i.e., non-Faradaic) responses (fractions of current flowing in response to the changing electrode potential) of an electrode in a CV, following a line of reasoning briefly reviewed elsewhere [3]. For further examples, see [106,107].

In a particularly popular and frequently encountered attempt, consideration starts at the power law relationship presented above Equation (7).

As stated, with $b = 1$, the current behaves capacitive-like. In a CV, further dependencies of current on scan rate can be observed. A case of particular interest is with $b = 1/2$. This is observed for the peak current in a cyclic voltammogram, both in cases where charge transfer and diffusion are limiting [108]. Assuming that the current response of an electrode can only be due to capacitive and faradaic contributions, it has been assumed

$$I = a \cdot v + c \cdot v^{1/2} \quad (16)$$

Such formally additive behavior was indicated first by Liu et al. [109] in a study of Mo_xN -electrodes. In a further stretch, it was assumed that this must be valid at all electrode potentials (although Equation (16) (the Randles-Ševčík equation simplified for the present discussion) was explicitly derived only for the peak current in a CV!). Rearrangement yields

$$I/v^{1/2} = a \cdot v^{1/2} + c \quad (17)$$

Plotting $I/v^{1/2}$ vs. $v^{1/2}$ yields slope a and intercept c . Liu et al. very logically concluded that the observed current must be subject to mixed control, i.e., caused by double layer charging as the capacitive process and some Faradaic reactions not further specified. Subsequently, this statement has developed an astonishing life of its own. It has been argued, presumably starting with a report by Lindström et al. [110], wherein the exponent in the power law equation (16) was observed to deviate from both 1 and $\frac{1}{2}$ considerably while being close to unity in an electrode potential region where hardly any Faradaic reaction could be expected. This region of electrode potential (an example can be seen above in **Figure 4**) was called, for good reason, for decades a double layer region. Subsequent authors (a brief overview has been provided by Ge et al. [3]) have assumed instead that there can be only 1 and $\frac{1}{2}$, with a and c being the adjustable parameters describing the Faradaic and the capacitive or pseudocapacitive parts of the current response. Evaluation of recorded CV data frequently resulted in displays of CVs showing major parts of the current (and charge) as being due to pseudocapacitive processes (some authors even simplified matters further, calling it the capacitive part). If this rather artificial separation makes any sense at all, it should suggest the conclusion that certain materials show higher fractions of the current response without being subject to diffusion limitation. For a material intended for a supercapacitor or a high-power battery, this is certainly a good outcome, but the same would have been obtained without all this rather diffuse reasoning by simply inspecting rate capability plots (capacitance vs. current density) showing a more constant capacitance or storage capability with a growing current density for a more suitable and structurally optimized material. Anyway, the above reasoning has been identified as lacking a scientific foundation [111].

The starting point of this reasoning hints at a development that again resembles a merger in the meaning of this report: to combine properties typical of a battery electrode and of a supercapacitor electrode.

The desire to obtain said separation for a single material, hopefully having significant amounts of both properties, whether it is of scientific and/or technological interest and relevance, appears to be a completely separate question that did not stop there. Various methods to obtain such separation have been compared; an example has been described by Forghani and Donne [112]. Three methods were compared (actually two, because two of the three methods are based on the same CV data, taking the I vs. v relationship (see above) and the integrated charge from a CV as separate methods) and resulted in the conclusion that the third method, named step potential electrochemical spectroscopy (SPECS) [113], presents the “most rigorous approach”. Given the fact that the current response after every electrode potential step was fitted using five adjustable parameters, this might not be too surprising. Somewhat surprisingly, electrochemical impedance measurements were not taken into consideration. This method, widely accessible with common instrumentation, also provides access to the interfacial (double layer) capacitance and the “capacitance” assigned to the redox process (the pseudocapacitance) [3,106,107].

Electrodes for both battery and supercapacitor applications are constructed with respect to performance (charge storage and current generation capability) and mass utilization [98]. Quite obviously, with a supercapacitor electrode, mostly the surface

and perhaps a few topmost layers are utilized in the electrode process, whereas in a battery electrode, in the ideal case, the whole volume up to the current collector is utilized in this reaction. Consequently, for the former application, a thin electrode will be preferred (because the bulk volume underneath the top layer will hardly be used, in particular in high-current applications), whereas for a battery electrode, a thicker volume with a smaller fraction of auxiliary material like a current collector is preferred. The transition from the former electrode (which has been called high power) to the latter (which has been called imprecisely high energy) is a continuous one, as depicted in the following **Figure 6**.

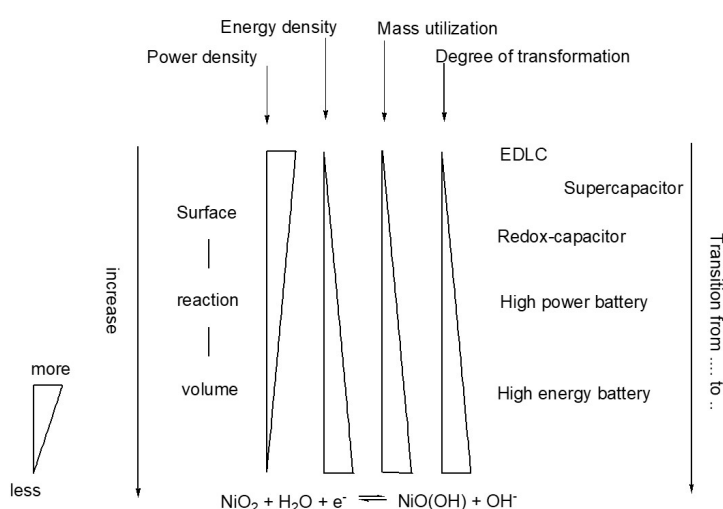


Figure 6. Transition from battery to supercapacitor electrode on the material and electrode design level.

Taking just this illustration, a supercapacitor electrode can be described as an extreme version of a high-power battery electrode. This “transition” was already addressed by Conway [43]. In **Figure 6**, the mode of energy storage, either by charge separation or by redox reaction, is missing because there is no conceivable continuum from charge separation to a redox reaction as proposed elsewhere [114]. Certainly, partial charge transfer can be misunderstood as a hint towards such a continuum, but close examination easily reveals that adsorption (most likely chemisorption) may include sharing charge between substrate and adsorbate, but in battery electrodes, no partial charge transfer happens.

Some further aspects and criteria can be added. A typical commercial example is Li-SOCl₂ primary batteries offered with a spirally wound electrode arrangement as a high-power battery and with a solid porous carbon core as the positive electrode and a comparably thicker lithium foil around it as the negative electrode in the high-energy version. Quite obviously, a cell and implicitly the employed electrodes meet both expectations: High energy and power are inherently impossible. Although scientifically not sound, studies of the relative fractions of capacitive and Faradaic currents addressed above have possibly contributed to the growing attention paid to optimized electrode structures and to material combinations enabling at least a closer approach to that elusive goal. An electrode material approaching this goal should have a large EASA, and this must be easily accessible for electrolyte solutions and electrolyte ions. To support high current, the material itself must provide high

electronic conductance, and the pores must not be too narrow to keep ionic conductance at a sufficiently high level. At this surface and very near it, a sufficient amount of battery-electrode material must be located to provide charge storage capability, finally resulting in the energy density of the complete cell. This description looks like a rather abstract wish list, but there is a simple test for the obtained achievement: Rate capability. A material successfully designed along the indicated lines of reasoning should provide excellent rate capability, even at high currents, and of course, only for short periods of time, the electrode potential for both electrodes should change hardly. The fundamental feasibility of this approach is illustrated by the lead acid battery and its remarkable power capability under discharge conditions (a researcher following the misleading line of reasoning briefly discussed above would assign a huge fraction of capacitive current to a lead acid battery during discharge). Unfortunately, during charging, this behavior was not found.

An approach working both during charge and discharge and reducing, in addition, the inventory of expensive electrode materials is the combination of RuO_2 nanoparticles and carbon materials with the metal oxide anchored on a foam of graphene and carbon nanotubes (CNTs) [115]. The obtained composite is stable, and the production is rather simple. The observed high energy density is based both on the sufficiently large surface area and the redox storage provided by the metal oxide. High rate capability is supported by the highly electron-conducting scaffold of the graphene foam with the attached CNTs. Unfortunately, no comparison with the state-of-the-art energy density of secondary batteries was provided, enabling a direct appreciation of the success of this approach.

The previous considerations can be assembled in a schematic approach (**Figure 7**) to an optimized electrode architecture valid both for battery and supercapacitor electrodes.

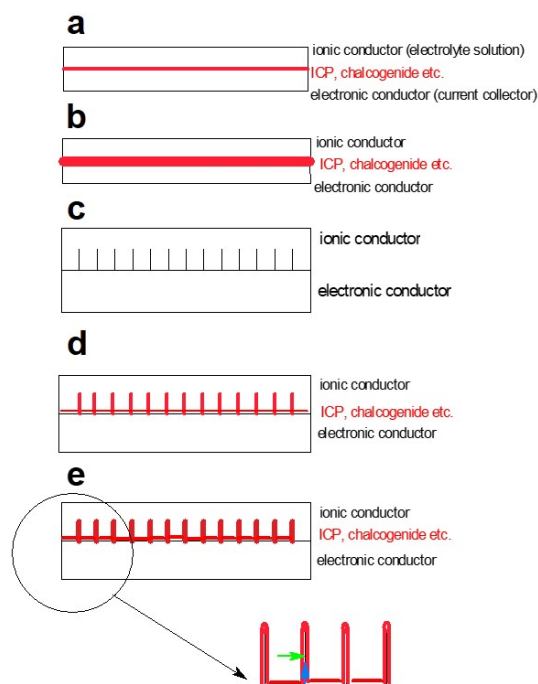


Figure 7. Sketches of electrode architectures.

Black: electronically conducting support; red: Active mass with ion (\rightarrow) and electron (\leftarrow) pathway.

A thin electrode (**Figure 7a**) may not provide enough active mass; a thicker, non-porous electrode (**Figure 7b**) has longer electronic pathways when the electrode reaction still takes place at the ICP/solution interface. An electronically conducting 3D support (**Figure 7c**) provides a larger interfacial area to be covered subsequently with active mass (**Figure 7d**), but the actual coating must finely balance electronic and ionic conduction pathways and their respective lengths and contributions to ohmic resistance (**Figure 7e** and insert). 3D-supports (instead of smooth metal foils) can be metal grids, foams or meshes, carbon or graphite paper, or carbon structures prepared by the pyrolysis of natural materials from biological sources. Further structuring on an even finer level can be afforded by controlled and directed ICP deposition. These considerations also apply to materials and electrodes for supercapacitors, and this has been highlighted before by Fu et al. [97].

5. Devices and cells

On the device or cell level, the merger proceeds in various ways. Electrodes containing both components acting via Faradaic reaction, like a conventional battery electrode, and acting via, e.g., a suitably large EASA [46], like a supercapacitor electrode, have already been discussed in the previous section. In addition, combinations of electrodes from both families will be the focus of this section. As already addressed above, the terminology appears to be confusing. A suggested distinction and proposed interpretation of the meaning of the terms “asymmetric” and “hybrid” have been reported [48]: An asymmetric device contains two electrodes of different materials but with the same storage mechanism, like a supercapacitor with different carbon electrodes, whereas a hybrid device contains electrodes with different modes of storage, like a negative activated carbon electrode (capacitive) and a metal chalcogenide positive electrode showing pseudocapacitive or battery electrode behavior. This distinction has been reaffirmed with numerous examples more recently [116].

The well-established limitations and flaws of the negative (lead) electrode in the lead-acid battery [117] have encouraged the search for alternative negative electrodes. Carbon-based materials were suggested, and the device also containing a conventional positive PbO_2 -electrode has been called a hybrid ultracapacitor [27]. The device just outlined can be called a capacitor (the rather confusing terminology of commercial devices¹ will not be followed here) or, provided its capacitance is large enough, a supercapacitor because the negative carbon electrode shows behavior typical of an EDLC-supercapacitor material. But if the intercalation of electrolyte ions proceeds, this may not be true anymore. The positive lead dioxide electrode behaves even less like a supercapacitor electrode; it shows battery electrode behavior [118]. Calling the device a battery because of this misses possibly a central property of the negative electrode. Attaching the term hybrid might help, but terms like pseudocapacitor (which even lacks basic logic) do not.

Starting at the same imperfection of the negative lead electrode, a device that might systematically be called a double hybrid—the authors prefer the term ultra-battery—with a positive PbO_2 -electrode and a negative one combining a lead and a carbon-based electrode into one has been proposed [119]. Simply connecting the

materials mechanically will not be enough. The electrode potential is now fixed (pinned) by the Pb(0)/Pb(II)-redox couple. As noticed, current vs. electrode potential relationships for the carbon electrode suggest a situation wherein during discharge, most of the current will initially be contributed by the lead electrode, with the carbon electrode contributing only towards the end [119]. During charging, things are slightly more as desired; initially, most of the current goes into the carbon electrode, but now, towards the end of charging, the negative carbon electrode will generate excessive hydrogen gas. Both situations are undesirable, but various modifications to electrode design and cell inventory nevertheless resulted in cell performance much better than that of state-of-the-art lead-acid batteries [119].

In an attempt to bring some order into the terminological confusion, Akinwolemiwa et al. tried to establish an extended terminology, including terms like negatrod and supercapattery of the second kind, etc. [35,13,103,120–123].

Instead of creating further terms and possibly adding confusion, some established terms and possible combinations are shown in **Figure 8**:

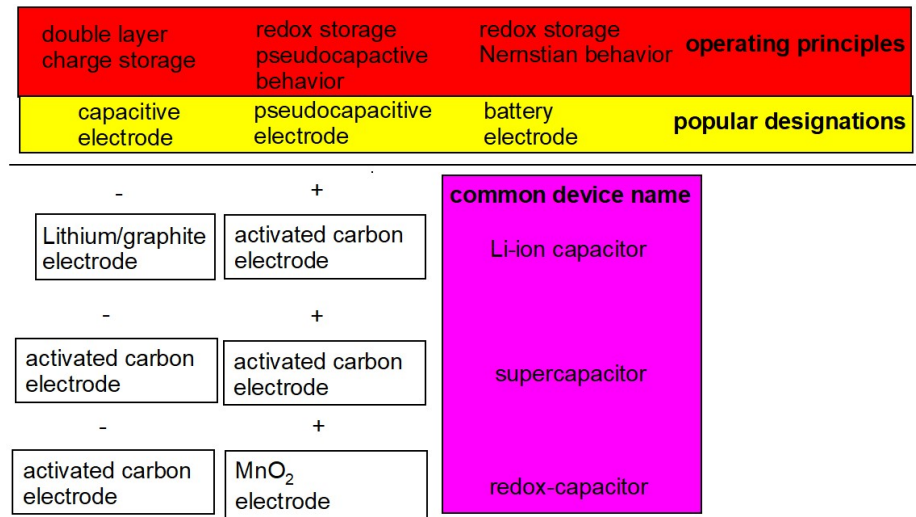


Figure 8. Possible combinations of electrodes and operating principles.

The term “Faradaic” is used to characterize a process involving charge transfer (different from non-Faradaic, i.e., capacitive processes); the term “Nernstian” is applied to a Faradaic process, showing the typical peaks observed in cyclic voltammograms. In this terminology, electrode processes showing a pseudocapacitive current response to a changing electrode potential may be called Faradaic and non-Nernstian, but as already suggested above and elsewhere [26], the creation of further names and terms as well as attempts to use terminology that is unnecessarily complicated appear to be of limited value only. For practical applications, it is of considerably higher interest to know how the charge/discharge behavior of a cell in terms of cell voltage vs. state of charge depends on the particular electrode combination. In the case of a capacitive electrode, there is no electrode reaction controlling the electrode potential; instead, the electrode potential linearly depends on the state of charge. In the case of a pseudocapacitive material, the situation is very similar, but the window of observed electrode potentials is controlled by the actual electrode reaction [26]. With a battery electrode, the actual electrode potential is

controlled by the activities of the participating reactants; basically, the potential will stay rather constant during the charge/discharge as long as a specific reaction proceeds. Only when the reaction switches to another one may the electrode potential show a noticeable step-like change (like in the case of the MnO_2 -electrode in an alkaline battery, wherein the transformation of MnO_2 proceeds in two different reactions associated with significantly different electrode potentials and correspondingly different cell voltages). Accordingly, the various combinations sketched in **Figure 8** will show different charge/discharge curves, basically somewhere between the capacitor- and the battery-type. These two boundary cases are illustrated in **Figure 9**.

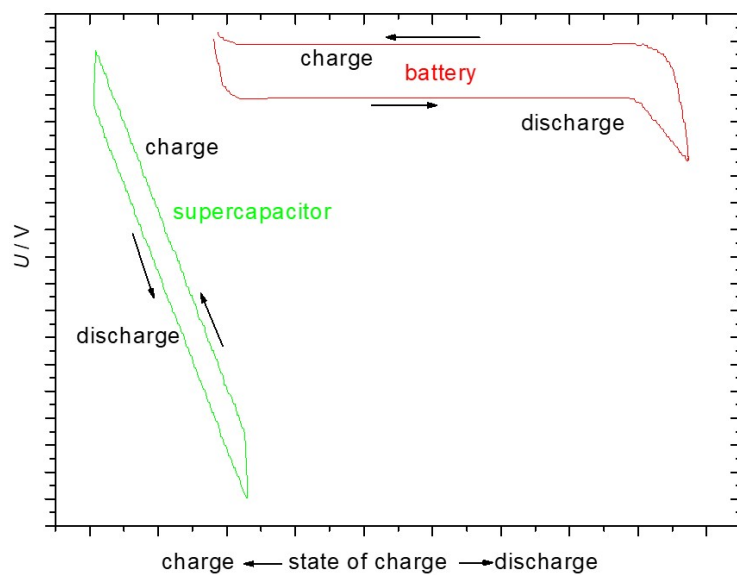


Figure 9. Simplified charge/discharge curves of a battery and a capacitor.

The actual shape of the curves will also depend on the matching of the storage capabilities of both electrodes.

Determination of performance data achieved for such devices and reporting with still missing or poorly defined standards enabling comparisons between more or less different systems was addressed frequently [3,15,116,124] with only limited success. Because the high cycling stability of supercapacitors, in particular the EDLC-type, is always stressed as a major advantage and a basic requisite for practical application devices placed between the traditional fields, whether hybrid or asymmetric, they should be examined with realistic parameters. Even when accepting the generally lower cycle numbers expected from batteries, a few hundred cycles observed with an electrode or even a complete system in this “in between” field is hardly useful.

Combinations of electrodes providing fast charge/discharge with limited storage capability with those showing larger storage capability but slower charge/discharge may look like a poor compromise because the power capability of the device will be limited by the electrode showing a smaller current capability, whereas energy storage is limited by the electrode having a lower charge storage capability. In large-scale and long-term energy storage (where most electrochemical energy conversion and storage systems are at a price disadvantage anyway), these concerns may be valid. But in applications where only short-term storage is required combined with, e.g., long-term stability, like in a power supply for an elevator where a supercapacitor may reduce the

peak power demand from 33 kW to 2.5 kW [125], this is no bad compromise anymore. Even the initial example of the carbon-lead dioxide system enjoys this benefit of high stability and fast charging because of the “fast” negative supercapacitor electrode and the “fast” positive electrode, which enable fast charging even during short periods of bright sunshine.

6. Conclusions

A growing number of electrode materials and architectures are encountered both in secondary batteries and redox supercapacitors. The operating principles, i.e., redox reactions, of such batteries and supercapacitors are basically the same; the major difference is the preferred location of electrode processes at the electrochemical interface in the latter and all over the electrode volume in the former. Consequently, scientists active in research and development in both fields should always look at the other side for inspiration and solutions. This applies in particular to the development of supercapacitor components aiming at higher energy densities, possibly to a lesser extent to high-power batteries. Obviously, progress achieved here can possibly be translated into improvements in batteries, possibly even for those who seem to be “adult” in terms of engineering.

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Notes

1. For supercapacitors no unified and universally accepted nomenclature has been established as of the time of writing. The terms supercap or supercapacitorTM (as well as ultracap/ultracapacitor) appear to lack a proper and generally accepted definition. At first glance it appears sufficient to assume, that capacitors based on the capacitive properties of the electrochemical double layer instead of a dielectric material like Al₂O₃ or Ta₂O₅ showing huge capacities are correctly called supercapacitors. Temporarily the latter term was trademarked (from August 1978 on) to NEC Corporation, currently this protection has apparently expired. The acronym SC seems to be too short to enable immediate identification. Acronyms like ES for electrochemical supercapacitor or FS for Faradaic supercapacitor do nothing beyond enlarging the confusion. Recently the device wherein purely electrostatic charge storage in the double layer is operative has been frequently called EDLC (electrochemical double layer capacitor). Thus it appears to be reasonable to call devices with high volumetric capacitance (in F), wherein charge storage is based both on electrostatic charge separation (like in an EDLC) and on Faradaic redox processes (including pseudocapacitive and redox storage) supercapacitors. Because of the combination of these fundamentally different charge storage mechanisms these devices are also sometimes called hybrids – adding further to the confusion. In the present report supercapacitors are such hybrid devices, the term ultracapacitor is not used at all. Its use to designate only those devices employing pseudocapacitances seems to be a losing proposition [A. Burke, J. Power

- Sources 91, 37 (2000).]. The statement, that B. Conway coined the term supercapacitor in 1991 is apparently erroneous. The rich collection of terms – some of them presumably protected by trademarks – does not help really: APowerCap, Best-Cap, BoostCap, CAP-XX, DLCAP, EneCapTen, EVerCAP, DynaCap, Faradcap, GreenCap, Goldcap, HY-CAP, Kapton capacitor, Super capacitor, SuperCap, PAS Capacitor, PowerStor, PseudoCap etc. Adding to this collection by defining the obvious like electrochemical supercapacitor with ES (all known supercapacitors are electrochemical devices, no others are known) or to interpret the well-known acronym EDLC by calling it an electrostatic super-capacitor as done by Wang et al.[6] does not help, either.
2. Recently the term pseudocapacitor has been used with growing frequency. As discussed before in detail this term is as wrong as the term pseudocapacitive for designating a material which does not behave pseudocapacitive at all [26, 48]. In short: From a linguistic and systematic point of view a pseudocapacitor must be a device behaving like a capacitor without being a capacitor. When accepting the certainly correct statement that a material like RuO₂ may show a capacitive-like response in a CV without being a purely double layer capacitor electrode material the term pseudocapacitor may be perhaps applied to a very narrow class of supercapacitors employing only such materials. But presumably misuse of the term would result in the same chaos currently observed with the term pseudocapacitive. Provisionally in this report the term redox-capacitor is used.
 3. The term dual-ion battery DIB stresses the fact, that two (both in case of a binary electrolyte) ions of the electrolyte participate in charge storage different from the rocking-chair principle of e.g. the lithium ion battery wherein only one ion participates. The acronyms DGB and DCB refer instead to the electrode material (G graphite, C carbon) missing the dual ion feature. DIC as dual ion cell is slightly more general.
 4. This as an extremely abbreviated notation. Of course the electrode itself is not capacitive, it is the electrode's behavior. This sounds like linguistic and possibly irrelevant hairsplitting, but the sometimes highly emotional, almost offensive discussion of this terminology suggests otherwise.

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