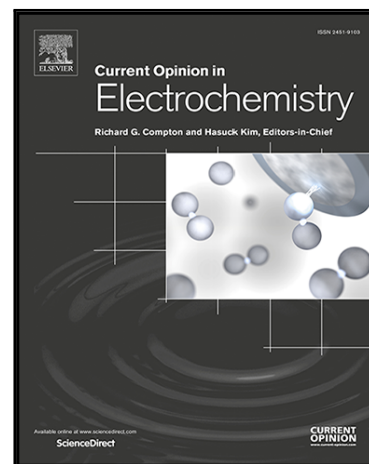


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Highlights

- The future of the EDLC's technology will depend on the successful realization of a new generation of EDLC's electrolytes
- The paradigm for the identification of electrolyte components need to be changed
- The understanding of the ion-ion and ion-solvent interaction appears of paramount importance
- The investigation about the impact of novel electrolytes on the EDLCs performance needs to be extended to all components (active and inactive) of these devices.

Non-aqueous electrolytes for electrochemical capacitors

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This review supplies an overview about the most recent work which has been carried out toward the realization of innovative non-aqueous electrolytes for high energy electrochemical capacitors (EDLCs). The solvents, conducting salts and ionic liquids proposed in the past years are discussed, and their impact on the EDLC performance is critically analyzed. Furthermore, the actual status of the research dedicated to the identification, characterization and application of novel electrolytes is analysed. The aim of this latter section is to supply indications about the most important aspects, which still need to be investigated in order to realize a novel generation of electrolytes suitable for the realization of high energy EDLCs.

State of the art electrolytes

Electrochemical double layer capacitors (EDLCs or supercapacitors) are nowadays considered the devices of choice for many high power applications. In these devices, the energy is physically stored at the interface between the electrodes and the electrolyte, through the formation of a double-layer [1-4]. Since the double layer formation can take place in a very short period (seconds or milliseconds), EDLCs can be charged and discharged in seconds (or less) and display high power ($10 \text{ kW}\cdot\text{kg}^{-1}$). Furthermore, since the efficiency of the charge-discharge process is very high, EDLCs display an outstanding cycle life (up to one million of cycles). The energy of these devices is in the order of $8\text{-}9 \text{ Wh}\cdot\text{kg}^{-1}$ [1-4].

Several studies indicate that if the energy of EDLC could be increased from the actual values to $15\text{-}20 \text{ Wh}\cdot\text{kg}^{-1}$, the number of applications in which these devices could be introduced would heavily increase, leading to a remarkable growth of their market size [1, 2]. Consequently, in the last years tremendous efforts have been made toward the realization of high energy EDLCs.

The energy of EDLCs can be described by the equation $E = \frac{1}{2}C \cdot V^2$, in which C is the electrode capacitance and V is the operative voltage of the EDLC. Considering this equation, it is obvious that the most convenient way to increase the energy of EDLCs is to increase their operative voltage, which is directly related to the stability (chemical and electrochemical) of the electrolyte [3, 5].

The largest part of commercial EDLCs contain electrolytes consisting of a mixture of ammonium salts (in most of the cases tetraethylammonium tetrafluoroborate, Et_4NBF_4) dissolved in organic solvents such as acetonitrile (ACN) or propylene carbonate (PC) [1-3]. These electrolytes display good transport properties and they

can be utilized in a rather broad temperature range. Their use enables the realization of devices with an operative voltage in the order of 2.8-3.0 V. If the operative voltage is increased, however, these electrolytes are not able to maintain the outstanding cycle stability mentioned above [2-4]. The cycle life of EDLCs is one the main strength of this technology and an increase of their energy should not be achieved at the expense of this feature. Therefore, the introduction of superior electrolytes is presently seen as a primacy for the future of this technology.

Novel electrolytes for EDLCs

The electrolyte of EDLCs needs to exhibit good transport properties, high thermal stability, large chemical and electrochemical stability, low flammability and low toxicity and, of course, it should also be cheap. It is also important to state that in EDLCs the electrolyte is an active material and, thus, the ion concentration within the electrolyte should be as high as possible. Bearing in mind the development of novel electrolytes, these aspects need to be taken into account [5-8].

In the past years, several studies have been dedicated to the development of innovative electrolytes, resulting in the proposal and evaluation of novel solvents and salts [4-5]. In the case of the solvents, large efforts have been dedicated to the identification of alternative nitriles and carbonates. In the case of the first class of compounds, solvents like adiponitrile (ADN) [9], butyronitrile (BTN) and 2-methylglutaronitrile (2-MGN) [10], have been investigated as replacement of ACN. These solvents have been tested in combination with conventional and alternative salts, and it has been shown that their use allows the realization of EDLCs with operative voltages above 3 V and a good stability at room temperature. However, since the electrolytes based on these solvents have higher viscosities and lower

conductivities compared to ACN, their use is limiting the power of EDLCs [2, 6]. In the case of carbonates, butylene carbonate (BC) was one of the first alternatives, which had been investigated. It has been reported that BC-based EDLCs work with operative voltages higher than 3 V, but overall they do not display an improved performance compared to PC-based EDLCs [11]. In parallel, also lithium-ion batteries (LIBs) electrolytes, which consist of mixtures of linear and cyclic carbonates, have been considered. EDLCs containing these electrolytes can operate at 3 V and display good performance [12]. However, the limited safety of these electrolytes represented a serious drawback for their introduction into commercial devices. In the attempt to realize high voltage EDLCs, also other categories of solvents have been investigated [2, 4, 5] and recently, utilizing a new identification strategy based on computational screening, which will be discussed more in detail in the following, our group proposed the use of 3-cyanopropionic acid methyl ester (CPAME). We showed that the use of this solvent permits the realization of high performance EDLCs with an operative voltage as high as 3.2 V [13, 14]. In the case of the conducting salts, quaternary ammonium-based, phosphonium-based cations and more recently pyrrolidinium-based cations have been proposed as alternative to the conventional Et_4N^+ -cation [2, 4, 5]. The anion bis(trifluoromethanesulfonyl)imide (TFSI⁻) has been the most investigated alternative to BF_4^- -anion [2, 4, 5]. Several salts based on these cations and anions have been used in combination with PC and ACN for the realization of alternative electrolytes [5, 15, 16]. These investigations indicated that a proper ion selection is essential for the realization of advanced salts and, also, that the salt-solvent interactions have a tremendous effect on the operative voltage of EDLCs [5].

Taking these results into account, the use of electrolytes containing alternative solvents and salts appears certainly as a promising strategy for the realization of high voltage EDLCs. Further improvements, however, are required to replace the conventional electrolytes with these alternatives.

Ionic liquid as electrolytes for EDLCs

Ionic liquids (ILs) can display good transport properties, high thermal stability and high electrochemical and chemical stability [17]. Moreover, because they are salts, their use allows the realization of solvent-free EDLCs [18, 19]. Due to these properties, which are matching all the requirements mentioned above, ILs have been intensively investigated as electrolytes for EDLCs in the last 10 years. ILs based on the cations pyrrolidinium, imidazolium, azepanium, sulfonium and cyclic sulfonium and based on the anions BF_4^- , PF_6^- , TFSI, bis(fluorosulfonyl)imide (FSI), dicyanamide (DCA^-), tricyanomethanide ($\text{C}(\text{CN})_3^-$), have been the most investigated [2, 4, 5, 17-19]. It has been shown that solvent-free IL-based EDLCs can operate at potentials higher than 3 V and also, that they are capable of operating in a wide temperature range [17-20]. However, the power of these devices appears in most of the cases substantially lower than that of conventional EDLCs [5, 6]. The ILs considered above belong to the family of aprotic ionic liquids. In the last years, in the attempt to improve the behaviour of IL-based EDLCs, also protic ionic liquids (PILs) have been proposed and tested. It has been shown that when dry PILs are used, the presence of the proton(s) on the cation of these ILs is limiting the operative voltage of EDLCs utilizing these electrolytes to 2.5 V. Therefore, even if PIL-based EDLCs display good cycling stability, their use does not appear particularly appealing for the

realization of high energy EDLCs [21]. In parallel, also other families of ILs have been evaluated, e.g. polymeric ionic liquids and ionogels [17, 22-24].

Very recently, also ILs containing redox active moieties have been suggested [25-30]. The addition of redox active groups to the anion and/or cation of the IL allows these electrolytes to provide capacitance via ion adsorption at the double layer, as well as, via electron transfer of the redox reaction of the introduced redox active species. Possible examples for redox active moieties in ILs are anthraquinone, TEMPO and organometallic compounds, i.e., ferrocene. After the pioneering work of Rochefort and co-workers [25-27], Fontaine and co-workers reported a biredox IL system functionalized with anthraquinone and TEMPO in 3-butyl-1-methyl imidazolium bis(trifluoromethylsulfonyl)imide ([BMIm][TFSI]) [28-30]. They showed that the use of this IL, allowing a simultaneous utilization of non-faradic and faradic storage processes, has a very positive impact on the electrode capacitance. Considering these results, redox ILs turn out as promising novel electrolytes for supercapacitors. Nevertheless, many aspects related to the use of these ILs, e.g. self-discharge and cycle life, need to be further clarified to really assess the advantages and the limits related to the use of this new class of ILs.

ILs are certainly promising electrolytes, but considering the results obtained so far, the development of novel ILs with improved transport properties, and reasonable costs, appears still to be needed.

Toward new electrolytes for high energy EDLCs

Although important results have been achieved in the last years, further efforts are needed to introduce a new generation of advanced electrolytes. These efforts

should focus on the identification, characterization and application of these electrolytes.

In the past years, the identification of novel electrolytes has been made using a “trial and error” approach, based on the chemical knowhow of the scientists. Due to the enormous amount of chemicals that could be potentially used as electrolytes for EDLCs, however, this search is experimentally difficult and very time consuming. Hence, it is necessary to rationalize the search-identification process as much as possible. Recently, with the aim to reach this challenging goal, our group proposed the use of a combined experimental-theoretical approach for the search of novel electrolyte components [10, 13, 14]. This approach is based on searching the complete known chemical space for new compound classes with advantageous combinations of viscosities, melting/flash/boiling points, electrochemical stability windows, pKas and ion solubilities, which can be computed at either SQM or DFT level and subsequent COSMOtherm calculations. Utilizing this innovative approach, it was possible to screen almost 70 million compounds and to identify commercially available compound such as CPAME, which have been never consider before as electrolytes for EDLCs. As mentioned above, CPAME is presently considered as a very promising electrolyte component for the realization of high energy EDLCs. Considering these results, this approach represents therefore a novel and powerful tool for the identification of new electrolyte components suitable for EDLCs. Nevertheless, the use of this tool has been only partially investigated and further investigations and refinements are required.

In order to design novel electrolytes, the use of advanced characterization techniques appears of crucial importance. In the last years, several studies have been dedicated to the use of in-situ techniques, and some important achievement have been

made. Batisse et al. were able to monitor the amount of gas produced by the decomposition of the electrolyte on the carbon surface in relation to the applied potential using mass spectrometry [31], while Kim et al., to investigate the decomposition gases, applied Raman spectroscopy [32]. In order to understand the electrode-electrolyte interplay Griffin et al. used in situ NMR and EQCM (Fig. 1). In their pioneering work, the authors were not only able to show that a huge proportion of anions stay at the negative electrode (as well as cations at the positive electrode), but also that the in-pore concentration of anions and cations differ hugely at the same voltage excursion [33]. Such in-situ techniques represent a powerful tool for the understanding of the electrolyte properties, and their use should be certainly intensified in the future to understand the interplay between electrolytes and EDLC materials (carbonaceous and non-carbonaceous).

Until now, the analysis of the impact of the novel electrolytes has been mainly dedicated on the active material's capacitance. Only few studies focused on the interplay between novel electrolyte and the inactive components of EDLCs. Varzi et al. investigated the stability of novel water-soluble binders in conventional electrolytes and in ionic liquids, showing that these alternative binders have a comparable electrochemical stability and enhanced mechanical resilience compared to conventional binders in non-conventional electrolytes [34-36]. In a recent work, our group showed that the understanding of the anodic dissolution processes taking place on the Al surface is of crucial importance for the realization of stable high voltage EDLCs [16, 37]. The magnitude of this dissolution process is strongly related to the solubility of the Al complexes, which are formed on the Al surface, on the solvent. The higher is the solubility of these complexes on the solvent, the higher is the magnitude of Al anodic dissolution. We showed that the design of novel electrolytes

for high voltage EDLCs need to be made taking into account this point and, also, that the use of alternative solvents, e.g. CPAME, improved the ability to suppress such detrimental processes compared to conventional ACN (Fig. 2). These studies clearly indicate that in order to gain a comprehensive understanding of the degradation processes occurring in high voltage EDLCs, the interplay between electrolyte and inactive components needs to be carefully considered. Therefore, it is necessary to intensify research efforts toward these interactions.

Conclusion and outlook

The future of the EDLC's technology will depend on the successful realization of a new generation of EDLC's electrolytes having a balanced set of properties and competitive price. In order to reach this challenging goal, the research dedicated to this topic need to be improved in several aspects. First, it will be necessary to change the paradigm of the identification of electrolyte components, making this process faster and more precise than it is at present. Secondly, further insight about the chemical-physical properties of novel electrolytes components need to be acquired. In this context, the understanding of the ion-ion and ion-solvent interaction appears of paramount importance. Finally, the investigation about the impact of novel electrolytes on the EDLCs performance needs to be extended to all components (active and inactive) of these devices. This latter study appears necessary to develop EDLCs with a cycle life comparable to that of the state-of-the-art devices, but with higher energy density.

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References

- [1] P. Simon, Y. Gogotsi, Materials for electrochemical capacitors, *Nature Materials*, 7 (2008) 845-854.
- [2] F. Béguin, V. Presser, A. Balducci, E. Frackowiak, Carbons and Electrolytes for Advanced Supercapacitors, *Advanced Materials*, 26 (2014) 2219-2251.
- [3] F. Béguin, E. Frackowiak, Supercapacitors Materials, Systems, and Applications, Wiley-VCH, Weinheim, 2013.
- [4] C. Zhong, Y. Deng, W. Hu, J. Qiao, L. Zhang, J. Zhang, A review of electrolyte materials and compositions for electrochemical supercapacitors, *Chem. Soc. Rev.*, 44 (2015) 7484-7539.
- [5] A. Balducci, Electrolytes for high voltage electrochemical double layer capacitors: A perspective article, *J. Power Sources*, 326 (2016) 534-540.
- [6] A. Brandt, A. Balducci, Theoretical and practical energy limitations of organic and ionic liquid-based electrolytes for high voltage electrochemical double layer capacitors, *J. Power Sources*, 250 (2014) 343-351.
- [7] J.P. Zheng, The limitations of energy density of battery/double-layer capacitor asymmetric cells, *J. Electrochem. Soc.*, 150 (2003) A484-A492.
- [8] J. Zheng, J. Huang, T. Jow, The limitations of energy density for electrochemical capacitors, *J. Electrochem. Soc.*, 144 (1997) 2026-2031.
- [9] A. Brandt, P. Isken, A. Lex-Balducci, A. Balducci, Adiponitrile-based electrochemical double layer capacitor, *J. Power Sources*, 204 (2012) 213-219.
- [10] C. Schütter, T. Husch, M. Korth, A. Balducci, Toward New Solvents for EDLCs: From Computational Screening to Electrochemical Validation, *The Journal of Physical Chemistry C*, 119 (2015) 13413-13424.
- [11] K. Chiba, T. Ueda, Y. Yamaguchi, Y. Oki, F. Saiki, K. Naoi, Electrolyte systems for high withstand voltage and Durability II. alkylated cyclic carbonates for electric double-layer capacitors, *J. Electrochem. Soc.*, 158 (2011) A1320-A1327.
- [12] A. Laheäär, A. Jänes, E. Lust, Electrochemical properties of carbide-derived carbon electrodes in non-aqueous electrolytes based on different Li-salts, *Electrochim. Acta*, 56 (2011) 9048-9055.
- [13] C. Schütter, S. Passerini, M. Korth, A. Balducci, Cyano Ester as Solvent for High Voltage Electrochemical Double Layer Capacitors, *Electrochimica Acta*, 224 (2017) 278-284.*

Utilizing a novel identification approach based on computational screening, the authors were able to identify a novel solvent, CPAME, suitable for the realization of stable high energy EDLCs.

- [14] C. Schütter, T. Husch, V. Viswanathan, S. Passerini, A. Balducci, M. Korth, Rational design of new electrolyte materials for electrochemical double layer capacitors, *Journal of Power Sources*, 326 (2016) 541-548.
- [15] S. Pohlmann, C. Ramirez-Castro, A. Balducci, The Influence of Conductive Salt Ion Selection on EDLC Electrolyte Characteristics and Carbon-Electrolyte Interaction, *J. Electrochem. Soc.*, 162 (2015) A5020-A5030.
- [16] J. Krummacher, C. Schütter, S. Passerini, A. Balducci, Characterization of Different Conductive Salts in ACN - Based Electrolytes for Electrochemical Double - Layer Capacitors, *ChemElectroChem*, 4 (2016) 353-361.
- [17] D.R. MacFarlane, N. Tachikawa, M. Forsyth, J.M. Pringle, P.C. Howlett, G.D. Elliott, J.H. Davis, M. Watanabe, P. Simon, C.A. Angell, Energy applications of ionic liquids, *Energy & Environmental Science*, 7 (2014) 232-250.
- [18] M. Salanne, Ionic Liquids for Supercapacitor Applications, *Top. Curr. Chem.*, 375 (2017) 63.
- [19] A. Brandt, S. Pohlmann, A. Varzi, A. Balducci, S. Passerini, Ionic liquids in supercapacitors, *MRS bulletin*, 38 (2013) 554-559.
- [20] R. Lin, P.-L. Taberna, S. Fantini, V. Presser, C.R. Pérez, F. Malbosc, N.L. Rupesinghe, K.B. Teo, Y. Gogotsi, P. Simon, Capacitive energy storage from - 50 to 100 C using an ionic liquid electrolyte, *The Journal of Physical Chemistry Letters*, 2 (2011) 2396-2401.
- [21] A. Brandt, J. Pires, M. Anouti, A. Balducci, An investigation about the cycling stability of supercapacitors containing protic ionic liquids as electrolyte components, *Electrochim. Acta*, 108 (2013) 226-231.
- [22] G.A. Tiruye, D. Muñoz-Torrero, J. Palma, M. Anderson, R. Marcilla, Performance of solid state supercapacitors based on polymer electrolytes containing different ionic liquids, *J. Power Sources*, 326 (2016) 560-568.
- [23] V. Chaudoy, F.T. Van, M. Deschamps, F. Ghamouss, Ionic liquids in a poly ethylene oxide cross-linked gel polymer as an electrolyte for electrical double layer capacitor, *J. Power Sources*, 342 (2017) 872-878.
- [24] M. Brachet, T. Brousse, J. Le Bideau, All solid-state symmetrical activated carbon electrochemical double layer capacitors designed with ionogel electrolyte, *ECS Electrochemistry Letters*, 3 (2014) A112-A115.
- [25] B. Gélinas, D. Rochefort, Synthesis and characterization of an electroactive ionic liquid based on the ferrocenylsulfonyl (trifluoromethylsulfonyl) imide anion, *Electrochim. Acta*, 162 (2015) 36-44.
- [26] H.J. Xie, B. Gélinas, D. Rochefort, Electrochemical and physicochemical properties of redox ionic liquids using electroactive anions: influence of alkylimidazolium chain length, *Electrochim. Acta*, 200 (2016) 283-289.
- [27] H.J. Xie, B. Gélinas, D. Rochefort, Redox-active electrolyte supercapacitors using electroactive ionic liquids, *Electrochem. Commun.*, 66 (2016) 42-45.
- [28] E. Mourad, L. Coustan, S.A. Freunberger, A. Mehdi, A. Vioux, F. Favier, O. Fontaine, Biredox ionic liquids: electrochemical investigation and impact of ion size on electron transfer, *Electrochim. Acta*, 206 (2016) 513-523.
- [29] E. Mourad, L. Coustan, P. Lannelongue, D. Zigah, A. Mehdi, A. Vioux, S.A. Freunberger, F. Favier, O. Fontaine, Biredox ionic liquids with solid-like redox density in the liquid state for high-energy supercapacitors, *Nature materials*, 16 (2017) 446.**

The authors provide a very thorough investigation of an IL system with redox active moieties attached to both anion and cation. High specific capacitance and cycling stability were reported for devices using redox active IL, making this approach of

combining double layer capacitance with faradaic redox reaction certainly promising.

[30] C. Bodin, E. Mourad, D. Zigah, S. Le Vot, S. Freunberger, F. Favier, O. Fontaine, Biredox ionic liquids: new opportunities toward high performance supercapacitors, *Faraday Discuss.*, 206 (2017) 393-404.

[31] N. Batisse, E. Raymundo-Piñero, Pulsed Electrochemical Mass Spectrometry for Operando Tracking of Interfacial Processes in Small-Time-Constant Electrochemical Devices such as Supercapacitors, *ACS applied materials & interfaces*, 9 (2017) 41224-41232.

[32] J. Kim, E. Kim, U. Lee, I. Lee, S. Han, H. Son, S. Yoon, Nondisruptive In Situ Raman Analysis for Gas Evolution in Commercial Supercapacitor Cells, *Electrochim. Acta*, 219 (2016) 447-452.

[33] J.M. Griffin, A.C. Forse, W.-Y. Tsai, P.-L. Taberna, P. Simon, C.P. Grey, In situ NMR and electrochemical quartz crystal microbalance techniques reveal the structure of the electrical double layer in supercapacitors, *Nature materials*, 14 (2015) 812.**
Combining two different in situ techniques, the authors were able to provide novel information about the dynamic of the double layer formation and, also, about the behavior of the ions of the electrolyte during this storage process.

[34] A. Varzi, A. Balducci, S. Passerini, Natural cellulose: a green alternative binder for high voltage electrochemical double layer capacitors containing ionic liquid-based electrolytes, *J. Electrochem. Soc.*, 161 (2014) A368-A375.

[35] A. Varzi, R. Raccichini, M. Marinaro, M. Wohlfahrt-Mehrens, S. Passerini, Probing the characteristics of casein as green binder for non-aqueous electrochemical double layer capacitors' electrodes, *J. Power Sources*, 326 (2016) 672-679.

[36] A. Varzi, S. Passerini, Enabling high areal capacitance in electrochemical double layer capacitors by means of the environmentally friendly starch binder, *J. Power Sources*, 300 (2015) 216-222.

[37] J. Krummacker, L.H. Heß, A. Balducci, Anodic Dissolution of Al Current Collectors in Unconventional Solvents for High Voltage Electrochemical Double - Layer Capacitors, *ChemSusChem*, 10 (2017) 4178-4189.

By investigating the influence of unconventional solvents on the anodic dissolution of the current collector, the authors provide novel informations about the impact of this degradation process on the cycle life of EDLC containing non-conventional electrolytes.

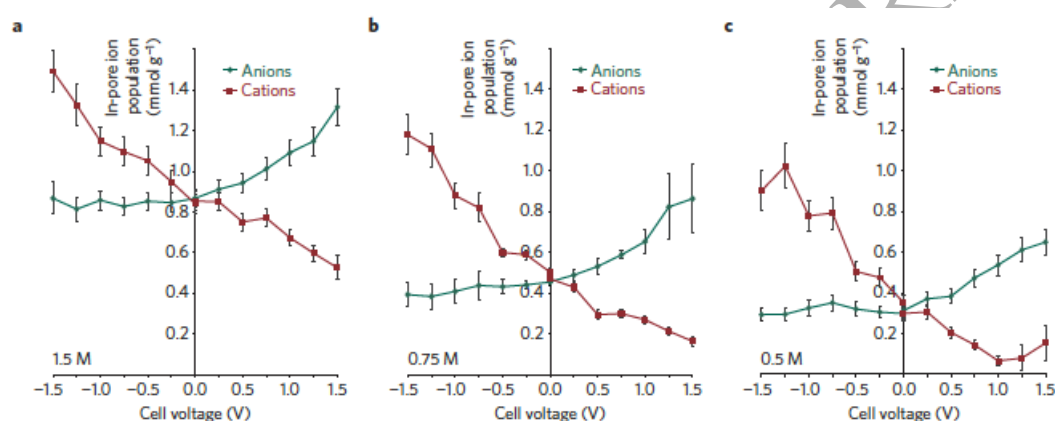


Figure 1: In-pore ion populations for supercapacitor electrodes at different states of charge in the range comprise between 1.5V to -1.5V. It is important to notice that the figure is reporting ion distribution with cell voltage and not with electrode potential. The figure a, b and c are relative to the in-pore ion concentration in electrolytes containing 1.5M(a), 0.75M(b) and 0.5M(c) of tetraethylphosphoniumtetrafluoroborate(PEt₄BF₄). Values are given as milimoles per gram of carbon in a single electrode. Error bars represent the range of values obtained from four independent datesets. Ion exchange is observed for positive charging whereas cation adsorption dominates for negative charging. Taken with permission from J.M. Griffin et al. [33]

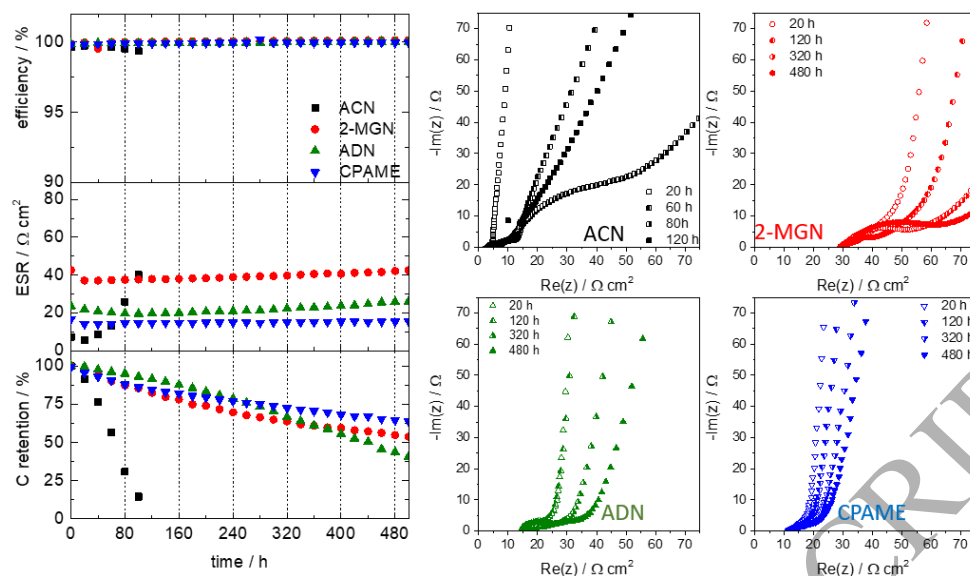


Figure 2. Comparison of the variation of efficiency, resistance, capacitance and impedance spectra of EDLCs containing alternative and innovative electrolytes (all with Et_4NBF_4) during float tests carried out at 3.1 V at RT. Taken with permission from J. Krummacher et al. [37]