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Lithium-ion batteries: Nomenclature of interphases with liquid or solid-state electrolytes

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Abstract

Although lithium-ion batteries are playing a paramount role in everyday life around the world, from portable electronics to electric vehicles, there seems to be no international organisation governing its development since the commercialisation began in 1991. As a consequence, the nomenclature of certain aspects of lithium-ion batteries are not clearly defined. For instance, no international consensus has been reached on the nomenclature of the interphases that play very crucial role in the operation of lithium-ion batteries. Unfortunately, this absence of proper nomenclature for interphases has been trending for far too long and it is confusing for emerging scientists, especially as it is being dragged to emerging technologies such as solid-state batteries and post-lithium chemistries. Here, the nomenclature problem of the interphases in lithium-ion batteries is critically addressed.

The present global technological advancements are such that there is not only the need for renewable or greener sources of electrical energy, but also a necessity for its effective storage and timely supply.^[1,2] Indeed, the indispensability of electrical energy storage (EES) is felt daily, and further concerted global effort is still required to fully satisfy this need. The development of efficient, affordable, and environmental-friendly electrochemical-energy-storage-systems is seen as the best option for a low carbon sustainable energy economy ^[3]. As shown in Figure 1, due to their outstanding energy density, Li-ion batteries are the preferred electrochemical energy storage systems. It can also be noted from Figure 1 that the gravimetric energy density of Li-ion batteries has seen an increase to 300 Wh/kg (as of 2022)^[4], a factor of approx. 4 in comparison to that first

reported in 1991 (~70 Wh/kg); there is a similar trend in volumetric energy density. It is worth emphasizing that the current improvement in energy density can be ascribed to advances in anode, cathode, and electrolyte materials as well as improvements in pack design.

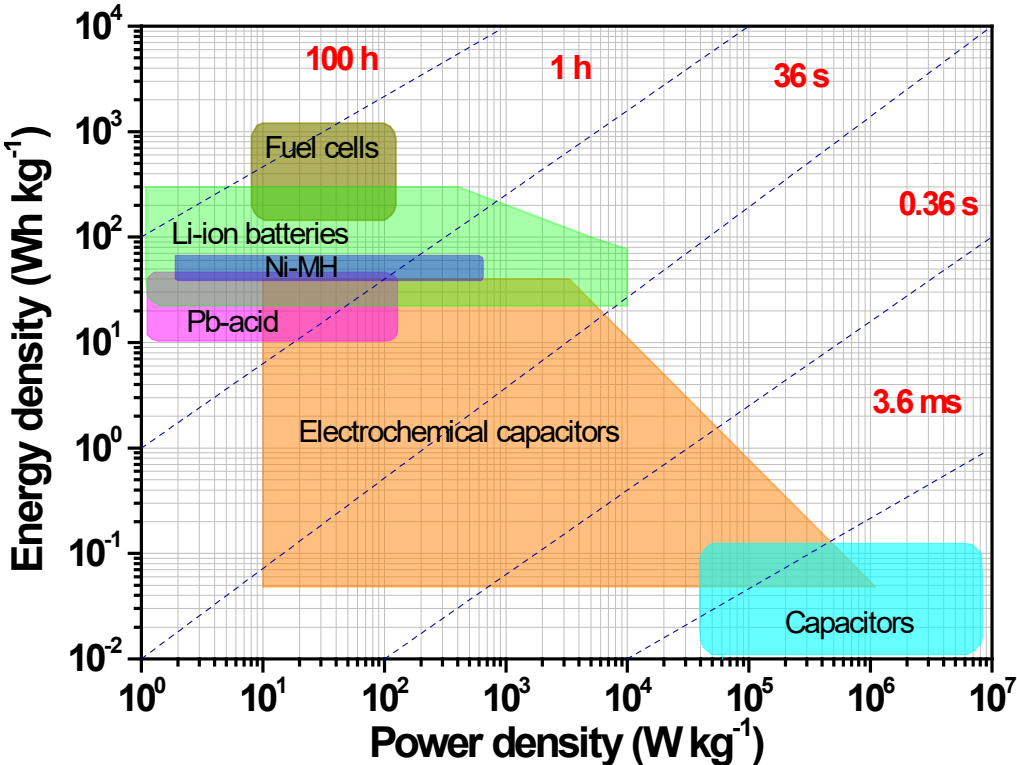


Figure 1. Ragone plot comparing the performances of various electrochemical energy storage systems and fuel cells (electrochemical energy generator). Typical charge/discharge times (runtimes) are demarcated by the blue dotted diagonal lines.

The commercialization of the Li-ion battery technology has been successful because of the extensive global research dedicated to its development over the past three decades. One aspect of the development that was particularly significant was the optimization of the interfacial films that form on the anode or cathode in contact with the liquid electrolyte. The interphases in a lithium-ion cell are almost insignificant in comparison to the overall size of the cell, but they play a paramount role in the operation of a lithium-ion cell – they are crucial in ensuring long lifetime; the typical thickness of the anode SEI in lithium and lithium-ion batteries is in the range of 10–100 nm^[5]. The carbonate-based electrolytes widely used in lithium-ion batteries go through reduction or oxidation reactions, on the surface of negative or positive electrodes, and form interfacial films on both electrodes, herein referred to as liquid-electrolyte/electrode interphase (*LEEI*) films. It is worth clarifying that an interphase refers to the interfacial film formed on the electrode, at the electrode/electrolyte interface or junction. The *LEEI* appellation is chosen herein

over the previously reported^[6] *EEL* (electrode/electrolyte interface) to reference the nature of the electrolyte involved.

As reported by Goodenough *et al*^[7]. in 2010, the difference between the Fermi energies of the electrodes and the HOMO/LUMO energy levels of the liquid electrolyte controls the thermodynamics and the driving force behind the formation of interfacial films on the positive and negative electrodes. When the LUMO level of the organic solvent or electrolyte lithium salt is lower than the Fermi level of the negative electrode, electrons will be injected into the LUMO orbit by that driving force, causing the electrolyte component to be reduced; when the HOMO level is higher than the Fermi energy level of the positive electrode, electrons are driven into the positive electrode, causing the electrolyte solvent or lithium salt to be oxidized^[7]. It is worth emphasizing that both the reduction and oxidation of the liquid electrolyte lead to the formation of interphases and consequently cause gases (C₂H₂, H₂, CO₂, etc.) to build up in the cell^[8]. More details about the SEI

These interfacial films that form at the anode and cathode can conduct lithium-ions and are at the same time electronically insulating, but critically, they must be chemically stable to prevent the electrode from further reacting with the electrolyte. If the interfacial film is not chemically stable, it will disintegrate/dissolve, and fresh interphases will keep forming and disintegrating till most of the liquid electrolyte in the lithium-ion cell is consumed. Such continuous formation/destruction of the interphase will lead to further gas build up, lithium inventory loss and the concomitant cell failure. Thanks to extensive global research efforts, various electrolyte additives have been developed that help to achieve very stable interfacial films on the electrode particles^[9,10]. Additives that are reduced just before the liquid electrolyte can help to prevent reduction of the liquid electrolyte at the anode; other additives are reduced together with the liquid electrolyte but enhance the integrity of the resulting interfacial film, making it robust.

The process of achieving stable interfacial films is called *formation*; this process is an important aspect of lithium-ion battery production^[11,12]. Although, originally focused on electrochemical formation of the interphase layers on the electrodes, the *formation* process has significantly evolved over the years to be more than simply electrochemical, comprising a combination of wetting or soaking conditions at varied temperatures for different durations and a series of electrochemical charge/discharge protocols^[11,12]. Though it has principally focused on the anode, the process is also believed to stabilize the interphase film on the cathode as well. The evidence of interfacial film on lithium metal anode was first reported by Peled in 1979^[13], it was

later confirmed for carbonaceous anodes thanks to the early work of Dahn and co-workers^[14]. On the other hand, the evidence of interfacial film on cathode materials was first reported by Thomas *et al.* in 1985^[15]. It is recommended to refer to previous publications^[5,16–19] for further information on the history and knowledge of interphase films in lithium-ion and post-lithium batteries. It is worth highlighting that interfacial films do form on cathode materials as well, even though most of them do not operate at potentials above 4.2 V vs Li⁺/Li. The nature of the interphase film at the anode has been studied in far more detail than that at the cathode^[6,20]. This is probably because the interfacial film at the anode is more delicate and has a more pronounced effect on the overall performance of the lithium-ion cell than that at the cathode, especially as the interphase film on the oxide cathode materials appears to be more robust than on the graphite anode.

To date, these interphases in lithium-ion batteries with liquid electrolytes are referred to as *SEI* (solid-electrolyte interphase) and *CEI* (cathode-electrolyte interphase) at the anode/electrolyte and cathode/electrolyte interfaces, respectively. The appellation *SEI* came about thanks to the work of E. Peled *et al.*^[13] in 1979, describing that the interfacial film formed on anode electrodes in contact with a non-aqueous liquid electrolyte behaved like a solid electrolyte as it conducted lithium ions, and, for some time now, the term *CEI* has emerged for the cathode interphase film – with no clear originator found yet. Edström *et al.*^[21] proposed the term *SPI* (solid permeable interface) for the cathode interphase film in 2004 but it was not adopted. So far, few publications have been found to have employed the term *CEI* as cathode electrolyte interphase film^[8,22–24]. To the best of our knowledge, it is suspected that the acronym *CEI* was proposed by Gallus *et al.*^[25] in 2015. Such *SEI* or *CEI* nomenclature for the interphase films on the anode and cathode, respectively, seem to be etymologically incorrect for lithium-ion batteries. It is incongruous that the anode interphase film is called *SEI* and the cathode interphase film is called *CEI*. Considering that similar degradation products (LiF, Li₂CO₃, ROCO₂Li, ROCO₂M, RO₂Li, MCO₃, MF₂ {M = transition metals}, polycarbonates, and polyethylene oxide) have been found in the interphase films on both negative and positive electrodes of conventional lithium-ion batteries^[6], they could have rather been given the same name: anodic *SEI* or cathodic *SEI* – unfortunately however, that has not been the case.

In fact, if the cathode/electrolyte interphase film is called *CEI* then the anode/electrolyte interphase film could also be called *AEI*; however, the *AEI* or *CEI* will not make any direct distinction if the interphase being referred to is with a liquid or a solid-state electrolyte. This misnomer is becoming increasingly significant, and must be addressed quickly especially as it is being dragged to emerging technologies like all-solid-state batteries. Hence, it is being proposed in this article that, with liquid electrolyte batteries, the anode/electrolyte and cathode/electrolyte

interphase films should be referred to as *LEAI* (liquid electrolyte/anode interphase), and *LECI* (liquid-electrolyte/cathode interphase), respectively. If oxide or polymeric coatings are deposited on electrodes for lithium-ion batteries to promote the stability of the liquid electrolyte in a cell, those coatings are *ex-situ LEAI* or *ex-situ LECI* rather than the *artificial SEI*^[5] that is circulating in literature.

Similarly, with solid-state electrolytes, the anode/electrolyte and cathode/electrolyte interphase films are proposed to be called *SEAI* (solid-electrolyte/anode interphase), and *SECI* (solid-electrolyte/cathode interphase), respectively. So far, it seems that the nomenclature for the interphases in all-solid-state batteries has also not been properly addressed yet. The state-of-the-art and proposed nomenclature are presented in Table 1.

Table 1. Summary of state-of-the-art and proposed nomenclature

	State-of-the-art nomenclature	Proposed nomenclature
General interphase in lithium or lithium-ion batteries	Electrode/electrolyte interface (EEI)	Liquid-electrolyte/electrode interphase (LEEI)
Interphase on anode with liquid electrolyte	Solid-electrolyte interphase (SEI)	Liquid-electrolyte/anode interphase (LEAI)
Interphase on cathode with liquid electrolyte	Cathode/electrolyte interphase (CEI)	Liquid-electrolyte/cathode interphase (LECI)
General interphase in solid-state lithium or lithium-ion batteries	None	Solid-electrolyte/electrode interphase (SEEI)
Interphase on anode with solid-state electrolyte	SEI or artificial SEI	Solid-electrolyte/anode interphase (SEAI)
Interphase on cathode with solid-state electrolyte	CEI or artificial CEI	Solid-electrolyte/cathode interphase (SECI)
Electrochemical conditioning of liquid-electrolyte cells after manufacture	Formation	None
Electrochemical conditioning of all-solid-state cells after manufacture	Formation	Initial coulombic cycling (ICC)

All-solid-state batteries are receiving a lot of development attention recently, with significant progress being made in solid electrolyte materials discovery and their processability^[26–29]. Figure 2 presents the room-temperature ionic conductivity band diagram comparing various solid-state electrolyte materials reported to date; the data for the plot was obtained from previous publications^[26,30–36]. Most of the solid-state electrolytes presented in Figure 2 have some stability issues either at lower or higher potentials and may require *SEEI* (solid-electrolyte/electrode interphase) films; that is, either *SEAI* or *SECI* films at particle level and/or electrode level for electrochemical stability. For example, the sulphides (Argyrodites, LISICON-types, etc.) are

known to form space-charge layers once in contact with cathodes^[37–39]. As a consequence, it is usually required for the cathode material particles to be given oxide coatings (called buffer layers) – via ALD or sol-gel techniques – in order to avoid the formation of space-charge layers and the concomitant interfacial resistance at the interface with the sulphide electrolyte^[37–39].

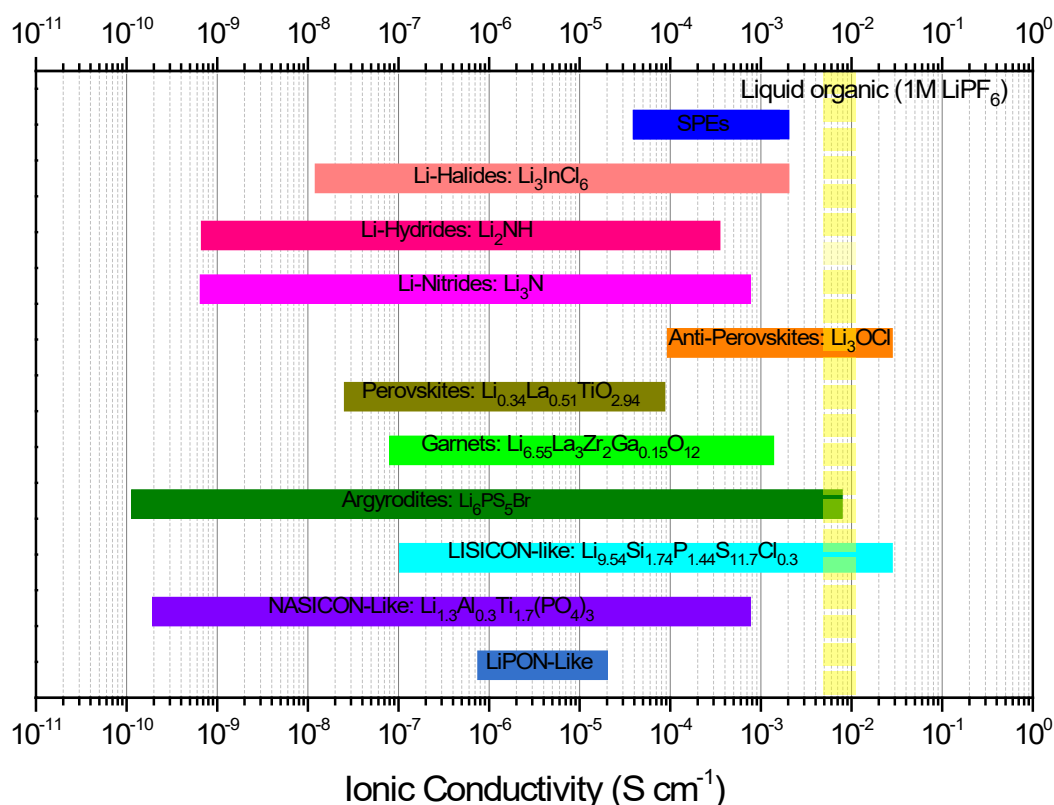


Figure 2. Room-temperature ionic conductivity band diagram for inorganic solid-state lithium-ion conductors and solid polymer electrolytes (SPEs). The chemical compositions given are just typical examples for each material type.

Such buffer layers are, in effect, pre-designed *SECI* films that allow the operation of the solid-state cell, just like the *LEEI* films (defined above) that allow the operation of the conventional liquid-electrolyte Li-ion cell. According to Koerver *et al.*^[40], despite the narrow thermodynamic phase field of some of the solid-state electrolyte materials, their practical use seems to be enabled by sufficient kinetic stability, i.e. the slow growth of interfacial films with reasonable ionic and low electronic conductivities. Thus, referring to such interphase films developed electrochemically in-situ or deposited ex-situ on the active materials – irrespective of whether the solid-state electrolyte is inorganic or polymeric – that allow the operation of solid-state cells as *SEAI* or *SECI* is more reasonable than the *SEI*^[41] or *artificial SEI*^[42–44] that are currently circulating for solid-state batteries. In fact, the interphase films formed on electrodes in liquid electrolyte cells have entirely different chemical nature or composition to what is deliberately deposited or electrochemically formed in solid-state cells. As a consequence, it will be reasonable to have their

corresponding nomenclatures. It seems the absence of an international global organization governing the development of batteries has led to such different terms or appellations circulating in literature, which can result in confusion, particularly for those new to the area.

What is more, the term *formation* also seems inapplicable to all-solid-state batteries, especially as there is no liquid electrolyte. Indeed, most of the *SEAI* or *SECI* films are deliberately deposited on the anode or cathode particles or at the electrode level, rather than being formed electrochemically during the first operation of the solid-state cell. Of course, there may be lithium loss due to reaction with surface impurities and/or adsorbed solvents, as well as lithium site loss due to structural irreversibility in the active materials that will reduce the first cycle efficiency. Though other interfacial films may inherently develop in a solid-state cell, especially without the *SEAI* or *SECI* films that are deliberately deposited to promote electrochemical stability^[40], the first electrochemical or conditioning cycles that occur in a solid-state cell until good coulombic efficiency is obtained is better referred to as *initial coulombic cycling (ICC)* rather than *formation*. The *ICC* process is also anticipated to be much faster (few hours) than the *formation* that occurs in liquid electrolyte cells during several days or weeks – which is notably, a significant cost in the production of liquid electrolyte cells, and may potentially be largely avoided for all-solid-state cells. These considerations of nomenclature are proposed to be applicable to conventional lithium-ion batteries as well as all-solid-state batteries with inorganic solid-state electrolytes (*ISSEs*) or solid polymer electrolytes (*SPE*) or their hybrids. They can also be applied to other related battery types, such as Na-ion batteries (or post-lithium chemistries), which are attracting growing interest, due to the potential for them to be inexpensive alternatives to lithium with more abundant raw materials for sustainability^[45,46].

The nomenclature or acronyms proposed herein are not meant to be definitive but worth considering for discussion in the community. Just as some consensus has been reached on shipping requirements for safety of lithium-ion batteries^[47], it is evident that a consensus on nomenclature in this specific field is also required, and an international organization – if not the *IEA* (International Energy Agency) – is needed to be formed for such purposes, as the battery energy storage industry and community continue to grow to meet the world's future energy storage requirements.

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