



Review article

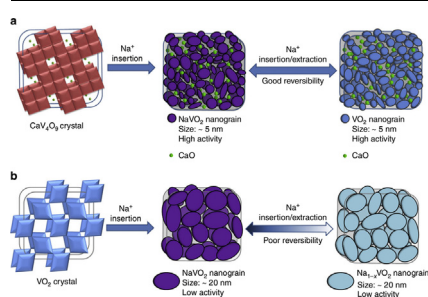
Sodium-ion batteries: New opportunities beyond energy storage by lithium

Ali Eftekhari^{a,*}, Dong-Won Kim^b^a Belfast Academy, 2 Queens Road, Belfast, BT3 9FG, United Kingdom^b Department of Chemical Engineering, Hanyang University, Seoul, 04763, Republic of Korea

HIGHLIGHTS

- Sodium-ion batteries are reviewed from an outlook of classic lithium-ion batteries.
- Realistic comparisons are made between the counterparts (LIBs and NIBs).
- The challenges and potentials of NIBs are subtly highlighted.
- NIBs need a subtle strategy of research and a pragmatic roadmap.

GRAPHICAL ABSTRACT



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ABSTRACT

Sodium has been recently attracted considerable attention as a promising charge carrier, but this sudden attention has made the strategy of research somewhat hazy, as most research reports are indeed the examination of typical materials rather than following a solid roadmap for developing practical cells. Although the history of sodium-ion batteries (NIBs) is as old as that of lithium-ion batteries (LIBs), the potential of NIB had been neglected for decades until recently. Most of the current electrode materials of NIBs have been previously examined in LIBs. Therefore, a better connection of these two sister energy storage systems can shed light on the possibilities for the pragmatic design of NIBs. The first step is to realise the fundamental differences between the kinetics and thermodynamics of Na as compared with those of Li. In fact, tiny differences between the electrochemical behaviours of these systems can lead us to new practical ideas for designing suitable materials. Furthermore, NIBs should be considered as new opportunities for energy storage rather than replacing LIBs. Hence, the subtle strategy of research is to learn from LIBs but not replicate them when designing NIBs.

1. Objective

1.1. Historical background

The history of sodium-ion batteries (NIBs) backs to the early days of lithium-ion batteries (LIBs) before commercial consideration of LIB, but sodium charge carrier lost the competition to its lithium rival because of better choices of intercalation materials for Li. During the 1960s, various electrochemical reactions were utilised for designing batteries, but most of these ideas did not survive for more than a few years. The

idea of LIBs in that era has been well reviewed by Scrosati [1] and Jasinski [2]. The same strategy was also followed in the 1970s, but some ideas became the centre of attention. For instance, metal-sulphur batteries were subject of numerous investigations. At the preliminary stage, the key requirement was to design a working system rather than maximising the specific energy by a lightweight charge carrier. Therefore, the focus was on sodium-sulphur battery [3] rather than the lithium-sulphur counterpart.

In the late 1970s, the boundary of solid state science and electrochemistry was indeed a hot topic due to the growing interest in ionic

* Corresponding author.

E-mail address: eftekhari@elchem.org (A. Eftekhari).

conductance in solid structures [4]. Thanks to the massive advancement in electrochemical instrumentation at that time, electrochemical insertion/extraction of ions could be carefully monitored to understand the mass transport within solids. Reversible insertion/extraction of Li and Na into/from transition metal dichalcogenides provided the basis for a new generation of rechargeable batteries [5–7]. Although this family of electroactive materials and even heavier members are still among the most common energy materials [8,9], the actual movement was made by the possibility of intercalation/deintercalation into/from layered metal oxides such as LiCoO_2 and NaCoO_2 [10]. It should be kept in mind that this merely represents the general trend, and the history of LIB is not straightforward. For instance, Campanella and Pistoia had reported the electrochemical intercalation of Li into/from MoO_3 in a non-aqueous electrolyte back in 1971 [11]. Earlier works employing a series of metal oxides have been reviewed by Jasinski [2,12]. However, the concept of intercalation/deintercalation was not a central area of research.

In any case, until the mid-1980s, the intercalation of alkali metals into new materials was an active subject of research considering both Li and Na somehow equally [5,13]. Then, the electrode materials showed practical potential, and the focus was shifted to the energy storage feature rather than a fundamental understanding of the intercalation phenomena. At this stage, due to better electrochemical performance of Li intercalation, Na became less attractive. Although Li could provide a higher specific capacity, the main motive for inclining towards Li was the excellent performance of electrode materials such as graphite anode and LiCoO_2 . Introducing the commercial prototype of LIB in 1991 by Sony was the last nail in the coffin of NIB. Since then, studies of NIB was a marginal area of research along with LIB. Despite the limited natural resources of lithium, there was no considerable scarcity in supply because the rapidly growing demand for LIB was limited to small portable devices. Nevertheless, the market demand for battery-powered electric vehicles provided an opportunity for the rebirth of NIBs.

There is now a tendency to catch up with LIB despite the lost history of NIB. The number of research publications on NIB may surpass that of LIB within a few years. This sudden attention can pave the path for the birth of new opportunities for the next generation of NIB (or more precisely, the first generation), but in the absence of a solid strategy of research, it is not easy to draw a roadmap. In the case of LIB, the central materials, graphite anode and LiCoO_2 cathode, were known from the early days, and the research strategies were to improve them or find alternatives. This is hardly the case for NIBs, as there is still no such central materials with satisfactory performance. Owing to the popularity of this area of research, numerous reviews have been recently published [14–28]. Most of these reviews provide a comprehensive overview of various materials, which have been examined for NIBs.

The present paper attempts to adopt a different strategy to provide an overall picture of the field. Instead of reviewing all available materials, the focus is on a selective collection from a conceptual perspective. These materials are compared with the Li counterparts to build a better outlook on the NIB status and potential. Many research papers consider both LIB and NIB together, but the purpose is usually to show the applicability of the material under consideration for both systems rather than a comparative study of the mechanistic differences. Furthermore, the distinguishable differences in energy efficiency, which is of both fundamental and practical importance [29], are highlighted throughout the paper.

1.2. Li vs Na: a realistic comparison

Owing to the rapidly growing interest in NIBs, it is common in the literature to compare NIBs with the available LIBs to highlight the advantages and disadvantages of NIBs. However, most of the statements are unrealistic and do not lead us very far. Therefore, it is of vital importance to clarify a realistic outlook for the future of NIBs. The key point is that a cell is composed of various components whose weights

and volumes are constant regardless of the charge carrier. Since the strategy of battery research is always practical to some extent, the theoretical values for capacity/energy should be 'ideally achievable' rather than superficial ones. A recent paper has quantitatively compared the maximum achievable values for various Li batteries and their counterparts [30].

When comparing LIB and NIB, two full cells should be compared. In the current cell architecture, only 2.5% of a LIB is made of the charge carrier, and the rest is the electrode materials, electrolyte, separators, current collectors, etc. Albeit, an excess amount of the charge carrier is required in the electrolyte, but the cell technology attempts to minimise it as it has been the case since the early LIBs. Since this is a theoretical comparison, we do not need to consider the availability of materials, and we can assume that graphite can accommodate Na to the same level of Li to reach the composition of NaC_6 and the NaCoO_2 can be charged to $\text{Na}_{0.5}\text{CoO}_2$ (or even assuming $\text{Na}_{0.5}\text{Li}_{0.5}\text{CoO}_2$ as the cathode material). In this case, the same amount of materials can be placed within the same volume since the volume of the Li and Na cells described above are almost the same (the densities of LiCoO_2 and NaCoO_2 are 4.9 and 5.0 g cm^{-3} , respectively). Therefore, the difference in the energy densities of the LIB and NIB is proportional to their operating voltage (*i.e.*, only 10% higher for LIB), and thus, the theoretical energy density of a NIB is only 10% lower than that of its Li counterpart. Note the terminology, when referring to NIB, the B stands for 'battery', which means at least a full cell.

It is often stated, even in the papers devoted to NIBs, that NIBs suffer from a low specific capacity as compared with LIBs. The comparison is indeed the theoretical capacities of metallic Li and Na anodes, though the available LIBs do not operate based on a metallic Li anode. The specific capacity of the anode has a minor impact on the overall specific capacity of the whole cell [30]. Theoretically, 1 mol of the charge carrier (7 g Li or 23 g Na) is required for 6 mol of carbon + 2 mol of $\text{Li}_{0.5}\text{CoO}_2$ (*i.e.*, 260.86 g of the active materials). As mentioned above, we can ideally assume that the discharged cathode materials are $\text{Li}_{0.5}\text{Li}_{0.5}\text{CoO}_2$ and $\text{Na}_{0.5}\text{Li}_{0.5}\text{CoO}_2$ for LIB and NIB, respectively. Hence, if calculating the specific capacity with respect to the weight of anode + cathode, the specific capacity of a NIB is only 10% lower than that of a LIB.

Considering 0.2 V lower cell voltage of a NIB as compared with a LIB, the overall theoretical specific energy is 279 Wh kg^{-1} and 231 Wh kg^{-1} for a LIB and a NIB [30]. Evidently, the target theoretical specific energy of NIB is only 18% lower than its LIB counterpart. Note that we did not even consider the weight of the electrolyte, outer shell, electronic components, additives, etc., which are constant weights regardless of the choice of charge carrier. Albeit, these are all theoretical considerations based on the constant weight of other components in accordance with the available 18650 LIBs. There are also possibilities for changing the cell architecture in favour of the specific energy, but these improvements can be made for either LIB or NIB, and there is no direct advantage for the lightweight charge carrier.

Theoretically, about 2% improvement in the specific energy is possible because of the opportunity of replacing the anode Cu current collector with a lightweight Al current collector in NIBs. This replacement also has a considerable impact on the cost too, as will be discussed in the next section.

1.3. Cost incentives

Despite the usual abundance of lighter elements, the natural resources for lithium are 1/1000 less than those of sodium. This has been initially considered as the prime motive for the development of NIBs, as it is still frequently repeated in the literature. For instance, a brief note calling lithium the new gold is among the highly cited papers of the field [31]. It is true that sodium is cheaper than lithium, but the cost of the charge carrier has a minor impact on the overall cost of a battery since the other components are more expensive, and a significant cost

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