



A short review of the effect of external pressure on the batteries

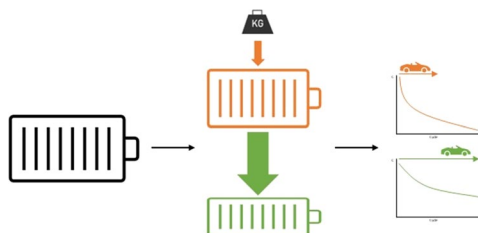
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Abstract

The research of the batteries is still going forward and there are lots of challenges which should be solved. This text examines the effect of external pressure on different types of batteries and explores their potential for improving performance and lifetime. The studies reviewed in the text show interesting results where external pressure affects capacity, internal resistance, stability or other parameters of modern battery systems as Li-ion, solid-state, or Li-S batteries. Despite the challenges, the benefits for next-generation batteries seem promising and show its role in battery development and manufacturing.

Graphical abstract



Keywords Silicon compounds · Solid-state · Electrochemistry · Ceramics

Introduction

In the context of global efforts to reduce emissions and achieve carbon neutrality, which are led by initiatives such as the EU's "Fit for 55" and the US Federal Sustainability Plan, scientists are actively exploring strategies to achieve these ambitious goals. One notable approach involves the widespread adoption of electric vehicles to reduce emissions from personal transport, as well as the use of electric heavy-duty vehicles to reduce emissions associated with transporting goods. As this transition proceeds, a large stock of lithium-ion batteries (see in Fig. 1) will enter the market and will become used [1, 2].

One research stream is focused on the research of the anode materials. The current generation of batteries utilizes two distinct types of anode materials. The predominant

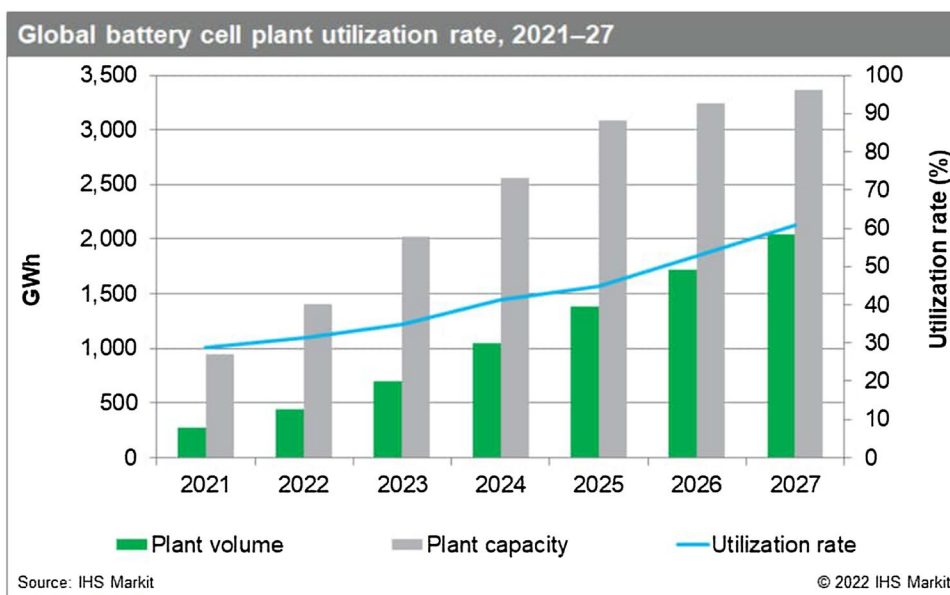
material employed by the majority of batteries is graphite. Graphite possesses a theoretical capacity of 372 mAh/g for LiC_6 and operates within a work potential range of 250 to 50 mV vs. Li/Li^+ [4, 5]. The second type of anode material employed is ceramic $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (LTO). LTO is utilized in use with LiMn_2O_4 (LMO) and $\text{LiNi}_{1-x-y}\text{Mn}_x\text{Co}_y\text{O}_2$ (NMC) cathode materials. The theoretical capacity of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ is 175 mAh/g [6], and it operates at a work potential of 1.55 V vs. Li/Li^+ [7, 8]. These materials show some volume changes during charging, these changes lead to faster degradation of the battery and in the worst cases can lead to battery failure [9]. In the case of graphite, LiC_6 volume changes are around 13% [10]. The LTO is more stable with volume changes lower than 0.1%, and because of that batteries with this anode have a high lifespan [11].

The present research focuses on strategies for enhancing the anode capability. Silicon has emerged as a promising material for augmenting the anode's capacity. $\text{Li}_{15}\text{Si}_4$ exhibits a theoretical capacity of 3579 mAh/g [12] and operates at a potential below 0.5 V vs. Li/Li^+ [7]. Capacity is ten times

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Fig. 1 Prediction of global production and production capacity of Li-ion batteries [3]



higher than today's commonly used graphite. Additionally, silicon possesses numerous active sites and has a minimal ecological footprint, it is the second most present element in the earth's crust (around 27%) [13]. Presently, batteries employ a Si/C composite, whereas silicon constitutes more than 5% [14]. However, it is anticipated that this proportion will rise by up to 40% in the future [12, 15]. In any case, several problems make it impossible to use this material. One of the biggest problems is volume changes during cycling up to 300% in comparison with conventionally used graphite LiC_6 13.2% [10]. This volume changes damage the SEI layer and its re-growing, which leads to capacity drop, another thing is that silicon particles crack and disintegrate which leads to more SEI layer growth [15].

In the case of next-generation solid-state batteries with Li metal as anode, there are problems like poor electric conduction made by low contact between electrolyte and electrode, low internal conduction made by solid electrolyte and lastly usage of bare Li metal, which grows inhomogeneously during charging and made lots of dendrites, which can affect safety [16–18].

Lithium-sulfur batteries (Li–S) are another promising successor of Li-ion batteries with their high energy density and high amount of cheap sulfur on the market. This technology is still in research due to several issues as low safety due to usage of bare Li metal and dendrite grow, low utilization efficiency of sulfur and its derivatives by its nonconductive behavior, dissolution of sulfur to the electrolyte and capacity fading caused by shuttle effect [19–21].

There are several ways of research in the battery field mostly focusing on Nex-gen Li-ion batteries and post-Li-ion batteries. New technologies struggle with several problems which impede practical use. Applying external pressure on

the batteries can solve some of these problems and significantly extend their lifespan by improving stability, suppressing the growth of internal structures, and enhancing energy efficiency. Therefore, further research is needed on how to improve the batteries and how to bring new improved batteries [22].

Because of the information mentioned above, studies dealing with the issue of using external pressure to enhance battery performance are presented in this article. This study can, therefore, serve as a reference point in current research focusing on the effect of external pressure on the physical electrochemical properties of advanced batteries. Result of the articles are summarized in Tables 1, 2.

Effect of the pressure on Li-ion batteries

There are several articles which focus on studying how external pressure affects the batteries. All these studies are focused on testing small pouch cells made in the laboratory. These studies mostly focus on reducing the negative behavior of next-gen Li-ion batteries with silicon anode, Li–S batteries, and Li-ion with solid-state electrolytes. Research is mostly made of pouch cells but in the case of solid-state batteries are typically used coin cells.

For example, the study made by Zhou et al. and his team focuses on comparing compressed and uncompressed NMC pouch cell batteries before and after ageing. It was observed that pressure has not had an effect on new batteries but after the ageing, the compression recovered partly lost capacity and after decompression did not lose increased capacity. Better results are achieved by lowering the internal resistance of compressed cells (see Fig. 2A) and recompressing the

Table 1 Summary of the results of the research for Li-ion batteries

System	Author	Focus of research	Results
Li-ion	Zhou [23]	Comparing compressed and uncompressed NMC cells before and after ageing	Capacity recovery; decrease of internal resistance after ageing
Li-ion	Koo [24]	Effect of external pressure on NMC pouch cell	3 MPa decreases the capacity and helps Li dendrites formation; 1 MPa better stability during cycling, and reduces dendrites growth
Li-ion	Choi [25]	Effect of pressure uniformity on pouch cells with Li anode using LFP and NMC cathode	Non-uniform pressure supported local dendrite growth; inactive lithium generated leads to a decrease of capacity and decomposition of electrolytes
Li-ion	Müller [27]	Prolonging the cycle life of batteries with silicon anode and managing swelling of the battery	With the right type of pressure, the swelling should be lower, efficiency is stable and the capacity lost should be lower
Li-ion	Bercmans [28]	Moderating 4 sizes of pressure on pouch cells with a silicon alloy anode	No significant difference between tested pressures but a significant difference with uncompressed. Increase in capacity, decrease of ohmic resistance and slower degradation. Negative effect on the shift between charging and discharging plateau
Li-ion	Göttlinger [29]	Testing the LTO electrode vs. Si electrode	After relithiating the LTO with 1 MPa coulombic efficiency of more than 99% at 1000 cycles, better structural stability

layers inside a battery [23], battery layers are decompressed during the ageing because of volume changes, gassing etc. (see Fig. 2B, C).

Koo et al. focused on the effect of external pressure on a single-layer NMC/graphite pouch cell with a capacity of 60 mAh. Pressures from 0 to 3 MPa were tested, and it was proven that at a pressure of 3 MPa, a significant decrease in capacity occurs with non-uniform growth of the SEI layer and significant Li dendrite formation. The optimal pressure of 1 MPa then enhanced the stability of the pouch cell during long-term cycling and reduced the growth of Li dendrites [25].

Choi et al. in their work focused on the effect of pressure uniformity on pouch cells with Li anode using LFP and NMC cathode. It has been revealed that the uniformity of pressure distribution is an essential parameter for Li anode pouch cells. An SP-PEEK-based holder was found to be the most suitable holder to provide this uniformity. Cell swelling is lower when uniform pressure is applied, and in the case of non-uniformity, local dendritic growth occurs at the areas of differential pressure. The inactive lithium generated leads to a decrease of capacity and decomposition of electrolytes [26].

The study of Müller et al. was focused on prolonging the cycle life of batteries with silicon anode and managing of swelling of the battery by flexible compression through spring and fixed by nuts and bolts. The results of this study showed that when is set the right type of pressure, the swelling should be lower, efficiency is stable and the capacity lost is lower [27].

Another external pressure test made by Bercmans et al. was focused on moderating four sizes of pressure on pouch

cells with a silicon alloy anode. Their result shows that there is no significant difference between these pressures, however, there is a significant difference in comparison with uncompressed battery. They achieve a 19% increase in capacity and a 50% decrease in ohmic resistance in discharge and lower degradation. The negative is a shift between charging and discharging plateau which leads to complications with measuring SoC [28].

Göttlinger et al. tested the LTO electrode vs. Si electrode which was stable without pressure and with pressure 0.2 and 1 MPa. With the pressure 1 MPa capacity decreases due to limited mass transport. After relithiating the LTO electrode with 1 MPa pressure, the battery was stable cycled and achieved 1000 cycles with coulombic efficiency ($99.63 \pm 1.07\%$). In addition, postmortem analysis shows big cracks formation in batteries without pressure, which leads to electrochemical inactive particles which leads to higher volume changes. In comparison, 1 MPa battery has smaller cracks [29].

Effect of the pressure on solid-state batteries

The effect of pressure is a widely studied area in solid electrolyte batteries, currently mainly in small-scale laboratory coin cells. The research team of Zhang et al. focused on the effect of external pressure on all-solid-state batteries. Their results show that external pressure helps with limited solid–solid contact (see Fig. 3) which increases ion transport. They were focused on inorganic solid electrolytes and organic solid electrolytes. In the case of inorganic electrolytes, external pressure increases the density and

Table 2 Summary of the results of the research for post Li-ion batteries

System	Author	Focus of research	Results
ASSB	Zhang [30]	Effect of external pressure on all-solid-state batteries	Increase ion transport; decrease dendrite growth; stable Li layer growth; external pressure prevents crack formation on anode
ASSB	Doux [31]	Pressure effect on sulfide electrolytes for solid-state batteries	50 MPa preparation pressure creates higher impedance at grain boundaries and lower cell activity. 370 MPa pressure improves capacity retention and rate capability, no significant effect on the cycling
ASSB	Doux [32]	Effect of external pressure to lithium plating/stripping on a symmetric Li cell with $\text{Li}_6\text{PS}_5\text{Cl}$ solid electrolyte	5 MPa allows stable and long-term lithium plating/stripping for more than 1000 h and stable cycling in a full cell
ASSB	Zhang [33]	The research focused on a mathematical model of the solid electrolyte–lithium interface	They concluded that pressures above 20 MPa are optimal in terms of sufficient contact and long-term lithium plating/stripping stability
ASSB	Sakka [34]	Use X-ray computer tomography to study the effect of the stack pressure on solid-state batteries	Increased stack pressure reduced the porosity, but enhanced the contact between solid electrolyte and electrode, which decreases charge transfer resistance and conductivity
Li–S	Schmidt [35]	Effect of external pressure on Li–S battery performance with two types of electrolytes (DME/DOL and SPSE HME/DOL)	External pressure increased the viscosity of the electrolyte and together with the effect on the electrodes, made it pressure denser with lower porosity. In the case of electrodes, external pressure has a positive effect on the Li anode, where reduced dendrites grow and make it more homogenous

ion conductivity of solid electrolytes by using high cold or hot pressing. In the case of organic polymer electrolytes, external pressure shows a negative effect on ion transport by folding and coiling of polymer chains. For lithium metal anode is recommended to apply lower pressures which can decrease the growth of dendrites and increase the stability of the Li layer growing. High pressure can push the Li metal through the solid electrolyte and cause a short circuit. With the anodes which have significant volume changes during cycling can external pressure prevent crack formations or detachment of electrodes. Their overall result is that external pressure has a significant role in all-solid-state battery performance and has a big impact on various aspects of the battery and its behaviour [30].

Doux et al. tested the pressure effect on sulfide electrolytes for solid-state batteries. In the research tested the usage of pressure during fabrication and in operation. Usage of pressure during fabrication has an effect on the overall performance of the battery. At lower production pressures (50 MPa), much porosity creates greater impedance at grain boundaries, which is detrimental to cell activity. Capacity retention and rate capability are significantly improved in batteries prepared at high fabrication pressure (370 MPa). On the other hand, they do not find a significant effect of the pressure on the battery stability during charging, so the battery does not need any special housing [31].

Doux et al. investigated the effect of external pressure to lithium plating/stripping on a symmetric Li vs. Li cell with $\text{Li}_6\text{PS}_5\text{Cl}$ solid electrolyte with the tested pressure varied from 5 to 75 MPa. It was found that a pressure of 5 MPa allows stable and long-term lithium plating/stripping for more than 1000 h and stable cycling in a full cell with an LNO-coated NCA cathode [32].

Zhang et al. focused their research on a mathematical model of the solid electrolyte–lithium interface and concluded that pressures above 20 MPa are optimal in terms of sufficient contact and long-term lithium plating/stripping stability [33].

Sakka et al. used X-ray computer tomography to study the effect of the stack pressure on solid-state batteries. They found that increased stack pressure reduced the porosity, but enhanced the contact between solid electrolyte and electrode, which decreases charge transfer resistance and conductivity. This leads to improved charge/discharge capacity. The best results were achieved in 50 MPa stack pressure. However, the further optimal pressure level must be studied [34].

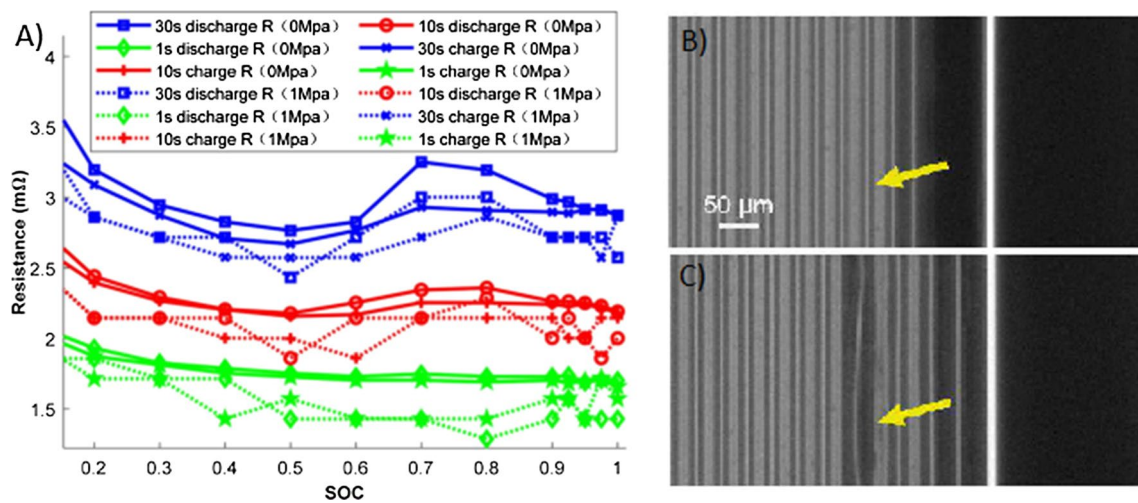


Fig. 2 A Comparison of internal resistance before and after pressure [5]. B Internal structure of cylindrical cell before ageing [6]. C Internal structure of cylindrical cell after ageing [6]

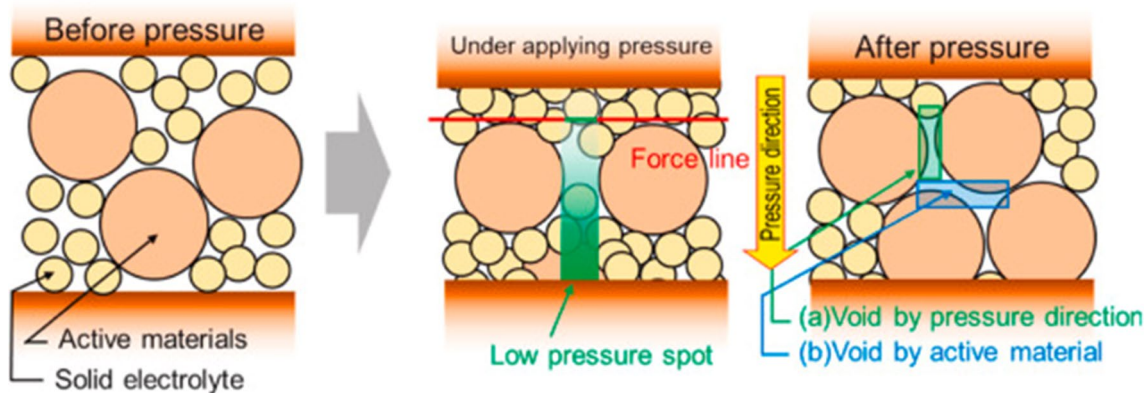


Fig. 3 Illustration of internal changes in an all-solid-state battery made by external pressure [10]

Effect of the pressure on Li-S batteries

The study made by Schmidt et al. was focused on the effect of external pressure on Li-S battery performance with two types of electrolytes: state-of-the-art DME/DOL (1,2-dimethoxyethane/1,3-dioxolane) and sparingly polysulfide solvating electrolyte (SPSE) HME/DOL (hexyl methyl ether/1,3-dioxolane). Battery filled with DME/DOL after applying external pressure showed low utilization of sulfur on the other hand battery filled with SPSE HME/DOL maintained a high utilization level even with high pressures. External pressure increased the viscosity of the electrolyte and in the combination of the effect on the electrodes, which made pressure denser with lower porosity, it can play a big role in battery performance. In the case of electrodes, external pressure has a positive effect on the lithium anode, where

reduced dendrites grow and make it more homogenous. The S/C composite cathode has pressure negative effect on porosity. Overall, the results show that with an appropriately chosen electrolyte and a suitably chosen pressure value, better results can be achieved in the production of high energy density Li-S batteries [35].

Conclusion

This text reviews the effect of external pressure on several types of batteries, including Li-ion batteries with silicon anode, Li-S batteries, and Li-ion batteries with solid-state electrolytes. These researches have shown that the controlled application of pressure can have a positive as well as a negative impact on battery performance depending on factors

such as the choice of materials, electrolytes and appropriate pressure levels. While in some instances, it can increase capacity, reduce internal resistance, improve battery stability and extend lifetime, it can also introduce problems such as worse charge transfer. Overall, the benefits of external pressure for the next-generation batteries outweigh its negatives and its applications need to be further researched.

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