A review of the use of chemical stabilisation methods for lithium-ion batteries

Mark D. Williams-Wynn[1] and Marcin H. Durski[1]

[1]EWaste Africa

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Abstract

The increasing amounts of end-of-life lithium-ion batteries (EOL LIBs) require novel and safe solutions allowing for the minimisation of the health and environmental hazards. Arguably, the best approach to the problem of EOL LIBs is recycling and recovery of the metals contained within the cells. This allows the diversion of the EOL battery cells from the environment but also the recovery of precious metals that can be reused in the manufacturing of new products, allowing to reduce the requirements of virgin materials from the mining industry. The most significant hindrance to the recycling process of EOL LIBs is their unstable chemical nature and significant safety hazards related to opening the air-tight casings. To minimise these issues, the end-of-life cells must be stabilised in one of the few available ways. This review aims at a comprehensive presentation of the studied chemical methods of EOL LIB cells discharge and stabilisation. The advantages and disadvantages of the method and its variations are discussed based on the literature published to date. The literature review found that a significant number of authors make use of chemical stabilisation techniques without proper comprehension of the associated risks. Many authors focus solely on the cheapest and fastest way to stop a cell from producing an electrical charge without extra thought given to the downstream recycling processes of safety hazards related to the proposed stabilisation method. Only a few studies highlighted the risks and problems associated with chemical stabilisation techniques.

Mark D. Williams-Wynn[1,2],Marcin H. Durski[1]*

[1] EWaste Africa, 102 Shortts Retreat Rd, Mkondeni, 3201, South Africa

[2] School of Engineering, University of KwaZulu-Natal, Howard College Campus, King George V Avenue, Durban, 4001

***Corresponding Author:** Email: mark@ewasteafrica.net, Tel.: +27 65 951 4836

Abstract

The increasing amounts of end-of-life lithium-ion batteries (EOL LIBs) require novel and safe solutions allowing for the minimisation of the health and environmental hazards. Arguably, the best approach to the problem of EOL LIBs is recycling and recovery of the metals contained within the cells. This allows the diversion of the EOL battery cells from the environment but also the recovery of precious metals that can be reused in the manufacturing of new products, allowing to reduce the requirements of virgin materials from the mining industry. The most significant hindrance to the recycling process of EOL LIBs is their unstable chemical nature and significant safety hazards related to opening the air-tight casings. To minimise these issues, the end-of-life cells must be stabilised in one of the few available ways. This review aims at a comprehensive presentation of the studied chemical methods of EOL LIB cells discharge and stabilisation. The advantages and disadvantages of the method and its variations are discussed based on the literature

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Keywords

Lithium-ion batteries, chemical stabilisation, over-discharge, recycling, industrial processes

Introduction

Secondary lithium-ion batteries (LIBs) are rechargeable electrochemical energy storage devices which have grown in popularity with advantages over other types of battery chemistries such as high energy capacities, long lifespans, resistance to self-discharge, and higher voltage output (Velázquez-Martinez, et al., 2019; Li, et al., 2018). As a testament to this, the LIB market has grown from 500 million cells produced in 2000 (Mossali, et al., 2020) to 4.5 billion cells produced in 2011 (Bernhart, 2014) to an estimated 25 billion cells becoming waste in 2020 (Yu, et al., 2018). Table 1.1 presents the generic compositions of lithium-ion batteries reported by Velázquez-Martinez et al. (2019) and Mossali et al. (2020). A cell comprises of anode and cathode materials separated by a porous material and enclosed in a sealed case (Figure 1.1). The case is filled with an electrolyte mixture to enable the movement of lithium (Li) ions between the electrodes of the cell to allow charging and discharging of the cell (Li, et al., 2018). In the charging process, Li^+ ions leave the cathode and move to the anode, where they intercalate between the graphite molecules (Sonoc, et al., 2015) as Li metal (Mossali, et al., 2020). The Li^+ions are unstable on the anode, and as the battery discharges, they move to the cathode, which creates an electrical current (Al-Thyabat, et al., 2013). A lithium-ion cell can only operate between approximately 1.5 and 4.2 V. At lower voltages, the copper current collectors become degraded, and at higher voltages, lithium forms reactive dendrites. Both phenomena can be detrimental to the safe operation of the cells (Mossali, et al., 2020).

Table 1.1. Lithium-ion cell construction components and materials according to Velázquez-Martinez et al. (2019) and Mossali et al. (2020).

Component	± wt. % (Velázquez-Martinez, et al. (2019))	± wt. % (Mossali, et al. (2020))	Commonly used mate
Case	25	20-26	Steel & plastics
Cathode	27	25-30	$LiCoO_2$, $LiNi_xMn_yCo$
Anode	17	15-17	Graphite or Li_4Ti_5O
Current Collectors	13	13-18	Cu or Al
Electrolyte	10	10-15	Solution of $LiPF_6$, Li
Separator	4	4-10	Microporous polyprop
Binder	4	2-4	Polyvinyl difluoride (

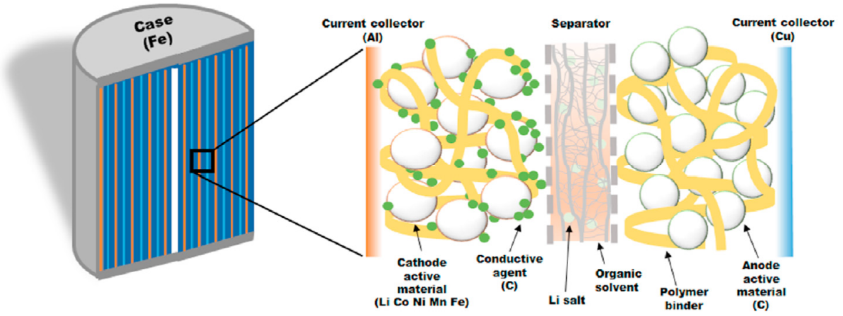

Figure 1.1. A schematic showing the construction of cylindrical LIB and the constituent elements used in each component (Kim, et al., 2021).

EOL Management

Most LIBs have a lifespan of between 3 and 8 years (Mossali, et al., 2020), after which they reach their so-called end-of-life (EOL). Individual cells from EOL LIBs, and particularly those from electric vehicles (EVs), can be reused for applications such as backup power and stationary energy storage units (Kim, et al., 2021). At present, this is feasible because the demand for used batteries far outweighs the supply of used batteries (Harper, et al., 2019). However, as the uptake of EVs increases, it is anticipated that the supply of cells (new and reused) will eventually outweigh the demand. At this point, recycling will become a necessity. Furthermore, a percentage of cells from EOL batteries cannot be reused due to irreversible damage that can occur. These have a final fate of recycling, regardless of the route they take to arrive there (Harper, et al., 2019). Currently, only a small percentage of the batteries that are produced are being recycled at EOL (Velázquez-Martinez, et al., 2019), with estimates that 95% of the LIBs that were produced globally in 2016 remaining stockpiled in old devices (Heelan, et al., 2016). This is predominantly due to most people not knowing how to safely manage EOL LIBs and being ignorant of the risks resulting from poor management (Mossali, et al., 2020).

Although recycling is a necessary treatment process for EOL LIBs, it is also a source of secondary raw materials (Velázquez-Martinez, et al., 2019; Harper, et al., 2019). Raw materials of particular interest from recycling include Co, Cu, Ni, Al, and Li, due to their high value (Li, et al., 2018; Zhang, et al., 2021). This is of importance due to the potential of demand outstripping supply for some materials, such as Li (Sonoc, et al., 2015). Additionally, in comparison to the production of virgin Li, in which ± 250 tons of feedstock are needed to produce 1 ton of Li, recycling can recover 1 ton of Li from ± 28 tons of feedstock (Harper, et al., 2019). Furthermore, the production of some virgin materials, such as Co, raise numerous social and ethical concerns (Harper, et al., 2019). More importantly, the recycling of LIBs prevents hazardous materials from entering the environment.

The two commonly used categories of LIB recycling are pyrometallurgical and hydrometallurgical processes (Kim, et al., 2021). Newer direct recycling processes are also under development, but these are not widely operated on an industrial scale yet. Pyrometallurgical processes are the most prevalent because they can handle large volumes of LIBs without need for pre-treatment and regardless of their state-of-charge (SOC) (Zheng, et al., 2018; Kim, et al., 2021; Rouhi, et al., 2021). When using pyrometallurgical processes, Ni, Co, and Cu are recovered from the batteries, but other materials such as the Li, Al and Mn are lost in the slag, while the graphite burns (Yi, et al., 2021). Hydrometallurgical processes are generally better equipped to enable the recovery of materials; however, they require shredding of the LIBs as a first step in the process (Wuschke, et al., 2019). The shredding processes must have a means of damping explosions and preventing fires or else additional stabilisation of the LIBs is required before shredding (Segura-Bailón, et al., 2024).

Discharging (stabilisation)

For LIBs, discharging process refers to processes where the amounts of metallic Li present in the cells are oxidised, preventing any violent reactions which occur when the Li is exposed to oxygen and/or water in the air (Duan, et al., 2022; Yang, et al., 2022). The discharging process (also referred to as stabilisation) forces the Li^+ ions to move from being absorbed by the graphite anode back to a stable cathode material. This means that lithium is thermodynamically stable and will not react violently if exposed to air or water (Punt, et al., 2022). This also allows to maximise the recovery of the lithium from the cathode by removing it from the anode. It also prevents short-circuits leading to thermal runaway (TR) events (Perea, et al., 2018; Sahraei, et al., 2012), as this would lead to the self-ignition of the metallic lithium present in the graphite anodes of the cells (Jena, et al., 2021; Zheng, et al., 2023) without the need for an external fuel source or oxygen once they have reached a certain temperature (Sommerville, et al., 2020)

Thermal runaway

Thermal runaway (TR) events in batteries can lead to major dangers, including the formation of hydrogen fluoride (HF) from the polyvinylidene fluoride (PVDF) binder, formation of carcinogenic nickel oxide, and exothermic decomposition of the conducting salt, $LiPF_6$ to form hydrogen fluoride (Hanisch, et al., 2015; Lee, et al., 2023). Nedjalkov et al. (2016) identified 11 different hazardous gases being released from a LIB while in TR. These include styrene, hydrogen fluoride and acrolein.

An oft-neglected fact is that the danger of TR exists even in stockpiled batteries awaiting processing (Harper, et al., 2019). It was reported that 50 % of all recycling and waste fires in the United Kingdom were due to stockpiled LIBs (Brown, et al., 2021). It is therefore important that these stockpiled LIBs be made safe to prevent TR. LIBs that are no longer susceptible to TR have the added advantage of significantly reduced transportation costs (Larouche, et al., 2020).

Stabilisation processes

There are several different techniques used for the stabilisation of LIBs. These techniques are listed in Table 1.2. There is limited research that has been undertaken on the stabilisation of LIBs. Of the 346 articles reviewed by Gao et al. (2024) only 75 even considered the discharge methods utilised as information worth mentioning, and even fewer of these gave more than a cursory mention of these discharge methods. This has led to an incomplete picture of the best methods of stabilisation (Fang, et al., 2022).

Table 1.2. Stabilisation techniques used for LIBs.

Stabilisation technique	Description
Electrical stabilisation	Electrical stabilisation methods remove the remaining energy that is stored in a batt
Chemical stabilisation	Chemical stabilisation methods use electrolyte solutions to short-circuit the battery,
Electrolyte removal	Removal of the electrolyte from the cells renders them inactive, as they can no longe
Mechanical stabilisation (in-process)	Mechanical stabilisation methods are also known as in-process stabilisation and cons

There is much debate about what a 'safe voltage' is in stabilisation processes and researchers have not reached any consensus on what is truly considered to be stabilisation of cells. For example,

- Peng et al. (2019) stabilised cells from spent LIBs to a residual voltage of less than 2 V,
- Marshall et al. (2020) discharged cells to below 2.5 V,
- Yao et al. (2020) considered a discharge to less than 0.5V as a full stabilisation, as a cell discharged to this level would release almost no further charge,
- Yao et al. (2020) also considered stabilisation to 1.0 V ("quick stabilisation") as sufficient to keep the dismantling process for LIBs relatively safe,

- Tao et al. (2023) discharged the cells to below 1 V,
- Lee et al. (2023) showed that significant swelling occurred in pouch batteries when discharged below 2 V, and they recommended that for safety, the cells should not be discharged below 2.5 V,
- Kwade & Diekmann (2018) recommended a discharge to a SOC of 0% (which corresponds to a voltage of less than 2 V),
- Wuschke et al. (2019) recommended a similar 2% SOC.

Once a cell has been stabilised to low voltages, it should not be recharged under any circumstances, as this could create safety hazards (Afroze, et al., 2023). One of the reasons for this danger is that the copper current collectors dissolve in the electrolyte once the voltage decreases to a certain level, and recharging results in the copper precipitating throughout the cell, resulting in short-circuits and TR (Harper, et al., 2019; Langer, et al., 2021; Hendricks, et al., 2020). Apart from TR, another concern with the deposition of Cu is the contamination of downstream products (Guo, et al., 2016).

Kaas et al. (2023) showed that for the most part, the discharge level of the batteries has no significant impact on the downstream mechanical comminution processes. They did find that there was greater copper dispersion for the cells that had their poles reversed through discharging, which correlates with the findings of Harper et al. (2019). Due to the impact that the discharge level has on the material composition, and therefore the downstream products, Kaas et al. recommended that additional studies into the optimal voltage to which the cells are discharged be undertaken.

Sonoc et al (2015) undertook a safety analysis of the hazards of large-scale dismantling of LIBs for recycling, and how stabilisation mitigates these risks. They opened several stabilised LIBs, with the cells that had only been stabilised to 0.5 V producing small red flames and fumes, but the cells that had been discharged to 0.0 V not only not producing any flames or fumes, but also not showing any evidence of reaction (temperature change or discolouration). However, Sommerville et al. (2020) contradicted these findings, as they showed that the cells from a large-format LMO-NMC battery were stable at 2.5 V when opened in air, and that these cells contained less than 2 % of the total energy that could be held at full charge.

Stabilisation compatibility with direct recycling and reuse

According to Lee et al. (2023), if direct recycling or reuse is intended, LIBs should not undergo stabilisation to 0 V. This is because this level of discharge can cause permanent damage to the LIBs, giving them a poor state-of-health (SOH) and potential swelling (Langer, et al., 2021). Swelling leads to damages to the cells, which would render the battery case, the cells, and the components useless for reuse. Instead, it was suggested that there is an optimum and appropriate discharge voltage that should be attained to keep the parts of the LIBs safe for reuse while preventing the risk of explosion or shock. The challenge with this is that the batteries would need to be manually dismantled while still holding charge. This would need to be done by operators with high-voltage training and insulated tools, or else by expensive robots (Harper, et al., 2019), which adds significant costs.

Battery & cell design

Most LIBs have some form of battery management system (BMS) with a cut-off low voltage. This cut-off voltage prevents the battery from stabilisation and becoming damaged. The various types of cell designs (cylindrical, prism, pouch, etc.) impact the hazardous nature of the cells as well as the best stabilisation technique (Harper, et al., 2019). Prismatic cells, for example, can withstand greater pressures and could be more hazardous in the case of a pressure build-up and rupture. Cylindrical cells, on the other hand, have positive and negative terminals at opposite ends, often with their own fuse. Furthermore, other aspects of battery design that could impact the ease of stabilisation, dismantling, and recycling include how the cells are joined; with this varying from nuts and bolts to welding and the use of potting compounds (Gaines, et al., 2018). Chemical stabilisation techniques are the most flexible for differing cell designs and for whole batteries without the need to disconnect from the BMS, as the terminals of the cells only need to be immersed in the liquid to ensure that the stabilisation can occur.

Safe transport of LIBs

Safety is a major concern in the transport of waste LIBs (Gaines, et al., 2018). Gaines et al. (Gaines, et al., 2018) report that, in the USA, air shipment of waste LIBs is no longer allowed, and rail shippers may also no longer accept EOL LIBs. With the rise of fires and explosions on ships over the last few years, and many of these attributed to LIBs, most shipping lines will also no longer accept EOL LIBs as cargo (NOW Media, 2022; Conway, 2023). As the fire hazard is generally only present so long as there is residual charge in the batteries (Rouhi, et al., 2021), stabilisation may be a means of enabling the shipping of EOL LIBs. However, currently, with the inability to ship EOL LIBs, small, local pre-treatment or recycling plants may need to be preferred.

Voltage Rebound or Relaxation

Voltage rebound or voltage relaxation is a phenomenon of voltage increase when a load is removed from the cell (Fuller , et al., 1994). It is usually studied as a useful phenomenon for extending the operational lifetime at voltages above 2.5 V and a SOC of greater than 0% (Rouhi, et al., 2021). The extent of the voltage rebound is dependent on both the temperature and the electrode materials used in the cells (Reichert, et al., 2013).

Voltage rebound means that the SOC of the battery undergoing stabilisation would be misrepresented, as the battery would contain residual energy even though the voltage readings suggest otherwise (Rouhi, et al., 2021). It is linked to the ohmic-transient behaviour of cells under load (Roscher & Sauer, 2011).

Methodology & Scope

This review of "chemical stabilisation techniques made use of in literature" searched virtual scientific literature databases with Google Scholar using the keyword phase, "LIB discharging recycling", with "LIB" also replaced with "lithium-ion battery". The search results were then screened to find articles that referred to chemical stabilisation techniques for LIBs prior to dismantling or recycling. In many instances, this reference to stabilisation was a single sentence referring to how the LIBs were prepared for the investigation being conducted. However, there were also several articles in which there was a substantial study on the chemical stabilisation process. The articles that made use of chemical stabilisation techniques are described in Section 3 of this study. Many stabilisation processes were only published in patents and not in academic literature due to the high economic importance of recycling business models (Piątek, et al., 2020; Sommerville, et al., 2020). There are also little data available on the stabilisation techniques used in industrial battery recycling processes. Information on the composition of the discharge solution utilised, the time allowed for stabilisation, or the results of the stabilisation are not readily available for these industrial processes in comparison to the data that are available for processes described for research activities.

Chemical stabilisation processes

There are two distinct uses of chemical stabilisation in literature: namely, the use of stabilisation to prepare batteries or cells for research activities, and the use of stabilisation as a pre-treatment for industrial recycling activities. However, in both cases the purpose is to prevent TR incidents, explosions and fires.

Research activities

In most research-based use of chemical stabilisation, the process is undertaken manually and on a small scale. There is often little consideration of the scaling of the process or the treatment of effluent. However, more details are provided on the composition of the discharge solution, the length of time of immersion and the temperature of operation.

Water as the discharge solution

Kang et al. (2010) placed LIBs in distilled water to discharge them after passing them through a roller press to contact the cathode and anode. These batteries were left in the water for a full day before removal, drying and downstream processing. Kim et al. (2014) also used distilled water as a discharge solution. According to Zhao et al. (2022), the immersion of charged batteries in water during the separation of the components is safe, efficient, and profitable. However, Sonoc et al. (2015) and Raj et al. (2022) pointed out that the difficulty and major danger associated with the stabilisation of LIB with pure water is the initial voltage of a charged cell will be higher than the electrolysis voltage of water, leading the splitting of the water molecules to produce oxygen and hydrogen gas These gases could lead to explosion risks if not handled safely with proper ventilation (Punt, et al., 2022).

Addition of salts

A salt, such as NaCl, is added to the water to avoid electrolysis of water in the chemical stabilisation of batteries (Hantanasirisakul & Sawangphruk, 2023) (Xiao, et al., 2020). The addition of the salt increases the electrolysis voltage of the solution and enables a faster discharge than can be achieved with water as the discharge solution. Even with the addition of salts, the discharge process can take more than 24 hours to complete (Or, et al., 2020). Although the addition of salts results in increased conductivity and electrolysis voltage, resulting in a more rapid discharge of the batteries, it also causes metal corrosion, which could result in the leakage of the electrolyte solution, leading to secondary pollution (Xiao, et al., 2020; Lee, et al., 2023). With the addition of NaCl, there is also a risk of chlorine gas generation (Kim, et al., 2021) with its associated toxicity risks (Sommerville, et al., 2020). NaCl solutions are the most used for stabilisation in research studies due to the high perceived discharge rate achievable and the low costs (Wu, et al., 2022). Other salts that have been proposed and investigated include $FeSO_{4(aq)}$ (Yao, et al., 2020), Na_2S (Torabian, et al., 2022), $MgSO_4$(Torabian, et al., 2022) and $MnSO_4$ (Xiao, et al., 2020). $FeSO_4$ is seen as a more environmentally friendly option, but it comes at a higher cost than NaCl. It also gives slower discharge rates, although Yao et al. (2020) showed that the performance of $FeSO_4$ solutions would be comparable to that of NaCl solutions if a cut-off safety voltage of 1V was used. $MnSO_4$ solutions give a slower discharge rate than NaCl solutions, but there is reduced galvanic corrosion, and this prevents the leakage of the electrolyte solution into the discharge solution (Xiao, et al., 2020). Most literature that mentions of the use of chemical stabilisation methods (see Table 3.1) give few specifics on how the stabilisation procedures were developed or tested (Ojanen, et al., 2018). As stated by Garg et al. (2024), much of "the literature concerning the electrochemical discharge of LIB was centred around the simplistic statement of: "batteries can be discharged in salt solutions" where the salt solutions were mainly NaCl or Na_2SO_4". A majority of the experimental papers in the literature do not investigate the performance of the specific salt that is used, and therefore are not able to comment on the suitability of the salt in comparison to water or other salt solutions. Only a few authors such as Ojanen et al. (2018), Shaw-Stewart et al. (2019), and Yao et al. (2020) have considered the impact that the salt used in the discharge solution has on the stabilisation process.

Table 3.1. Literature data on the solutions used and time of the chemical stabilisation reported

Author	Solution Used
Abdollahifar et al. (2023)	NaCl
Amalia et al. (2023)	10 wt. % NaCl, 10 wt. % NaOH
Anwani, et al. (2020)	10 wt. % NaCl

Author	Solution Used
Bi, et al. (2020)	5 wt. % NaCl
Chen, et al. (2018)	10 wt. % NaCl
Chen, et al. (2021)	0.5 M $Ca(Ac)_2$
dos Santos, et al. (2021)	0.1 M NaCl 0.2 M $MgSO_4$*7H_2O
Fan, et al. (2020)	NaCl
Fang, et al. (2022)	0.8 M NaCl, 0. 8 M $MnSO_4$, 0.8 M $FeSO_4$, 0.8 M KAc, 0.8 M $Zn(Ac)_2$
Fathima et al. (2024)	10 wt. % brine
Fu, et al. (2021)	10 wt. % NaCl
Guan, et al. (2017)	NaCl
He, et al. (2015)	5 wt. % NaCl
He, et al. (2017)	5 wt. % NaCl
He et al. (2017b) ()	5 wt. % NaCl
Huang, et al. (2018)	5 wt. % NaCl
Jafari, et al. (2020)	5 wt. % NaCl
Jena, et al. (2024)	5 wt. % NaCl
Li, et al. (2016a)	5 wt. % NaCl
Li, et al. (2016b)	Pure water, 5, 10 and 20 wt. % NaCl
Li, et al. (2018)	Saturated Na_2SO_4
Li, et al. (2019)	NaCl
Liu et al. (2024)	5 M NaCl
Lu, et al. (2013)	1, 5 and 10 wt. % NaCl
Lu, et al. (2019)	1, 5 and 10 wt. % NaCl
Mahandra & Ghahreman (2021)	1.0 M NaCl
Natarajan, et al. (2020)	NaCl
Nie, et al. (2015)	Saturated Na_2SO_4 with iron powder
Nie et al. (2023)	5.0 wt. % NaCl
Novaes et al. (2023)	0.1, 0.5 and 1.0 M $MnSO_4$
Ojanen, et al. (2018)	5, 10 and 20 wt. % solutions of NaCl, $FeSO_4$ and $ZnSO_4$ without stirring. 5 and 10 wt.
Pindar & Dhawan (2020)	1 wt. % NaCl
Pražanová et al. (2024)	5 to 30 wt. % solutions of NaCl, NaOH and $NaNO_3$
Punt, et al. (2022)	5 wt. % NaCl
Ra & Han (2006)	NaCl (brine)
Rouhi, et al. (2021)	5 or 10 wt. % Na_2CO_3 and K_2CO_3
Rouhi, et al. (2022)	5, 10 and 15 wt. % NaCl and $(NH_4)_2CO_3$
Sattar, et al. (2019)	NaCl (brine)
Segura-Bailón et al. (2024)	1M NaCl, Na_2CO_3 and NaOH solutions
Shaw-Stewart, et al. (2019)	5 wt. % NaCl, $NaHSO_4$, Na_2SO_4, $Na_2S_2O_3$, $NaNO_2$, $NaNO_3$, Na_2CO_3, $NaHCO_3$, NaOH
Sloop, et al. (2020)	Na_2CO_3(brine)
Song, et al. (2015)	0.8 M $MnSO_4$ and 2 g/l ascorbic acid
Torabian, et al. (2022)	NaCl, Na_2S and $MgSO_4$ at 12, 16 and 20 wt. %
Wang, et al. (2012)	NaCl
Wang, et al. (2018)	5 wt. % NaCl
Wang, et al. (2024)	5 wt. % NaCl Graphite medium
Wei et al. (2023)	5 wt. % NaCl
Wei et al. (2025)	10 wt. % NaCl
Wu et al. (2025)	1 M NaCl
Xiao, et al. (2017)	5 wt. % NaCl
Xiao, et al. (2020)	1 M NaCl, 1 M KCl, 1 M $NaNO_3$, 1 M $MnSO_4$ 1 M $MgSO_4$, 2 M NaCl
Xu, et al. (2008)	NaCl
Yang, et al. (2018)	5 wt. % NaCl

Author	Solution Used
Yang, et al. (2019)	NaCl (saltwater)
Yang et al. (2025)	10 wt. % NaCl
Yao, et al. (2018)	10 wt. % NaCl
Yao, et al. (2020)	0.8 M NaCl, $FeSO_4$ or $MnSO_4$
Zhang, et al. (2013)	5 wt. % NaCl
Zhang, et al. (2014)	5 wt. % NaCl
Zhang, et al. (2018)	5 wt. % NaCl
Zhong, et al. (2019)	5 wt. % NaCl
Zhong, et al. (2020)	Salt solution
Zhu, et al. (2021)	5 wt.% NaCl

Karli (2021) investigated the optimum temperature ranges for the stabilisation of prismatic cells to improve the recovery and quality of the cathode material from spent batteries. They found that slightly elevated temperatures of between (318.15 and 323.15) K were optimum for stabilisation using a salt discharge solution. According to Al-Thyabat et al. (2013), the time needed to achieve a sufficient stabilisation is dependent on several process parameters, including the conductivity of the solution, the temperature of the solution, and the state of charge of the LIB. There are therefore several variables that must be considered in developing a chemical stabilisation process. According to Harper et al. (2019), the stabilisation of batteries in salt solutions is not suitable for high voltage modules and packs due to the high rate of electrolysis and evolution of gases that would occur. It is however the cheapest in terms of operational costs (Sommerville, et al., 2020).

Although for the most part, the objective of chemical stabilisation is solely to reduce the voltage of the cells, there has also been some research into using the process to produce valuable materials. Rouhi et al. (2022) pointed to the electrolysis of ammonium-based solutions as a means of discharging batteries, as solutions such as ammonium carbonate and ammonium hydroxide were reported by Lu et al. (2019) to be less severe than other solutions. The process of electrolysis of ammonium salt solutions was envisioned by Boggs and Botte (2009) to produce hydrogen for fuel cells.

Multicomponent & complex solutions

Ali et al. (2022) suggested the use of ascorbic acid as an accelerant for the discharge process. This was also explored by Song, et al. (2015). The oxidation reaction that takes place with the ascorbic acid is anticipated to accelerate the discharge process. However, the ascorbic acid cannot be regenerated, making its use costly. For this reason, it has not been explored further.

Authors such as Ojanen et al. (2018) have proposed the use of a complex discharge solution containing solids rather than a simple salt solution. They showed that the addition of iron or zinc powder to the discharge solution could accelerate the discharge by up to 90%. Nan et al. (2005) placed EOL LIB into a stainless-steel container which was filled with water and 'electric iron powder'. The solution was then stirred, and the batteries were discharged completely after 30 minutes. The biggest challenge with this is the sacrificial oxidation of the powder, increasing the operational costs.

Garg et al. (2024) used a Fe(II)-Fe(III) redox couple electrolyte (more specifically, a 5 wt. % solution of potassium hexacyanoferrate and potassium hexacyanoferrate) for the chemical stabilisation of LIB to a voltage of 2.0 V after a rebound had occurred. This method ensured that there was no corrosion of the battery casing observed, ensuring that there was no leakage of electrolyte into the discharge solution. Gas evolution was still, however, observed in this discharging process, and this was attributed to the electrolysis of water molecules and the formation of H_2 and O_2. To get the voltage to below 2.0 V, a periodic discharge regime was necessary, as the voltage did not drop below 2.4 V with a single immersion despite immersion for up to 167 hours.

Mikita et al. (2024) proposed a so-called redox shuttle (RS) in the form of ferrocene and phenothiazine solution introduced into the spent cell through a small bore in the shell. The potentials of redox reactions induced by the RS must be between the positive and negative electrode potentials. The positive electrode is then reduced with electron acceptance from the RS and lithium acceptance from the electrolyte solution. In parallel, the negative electrode is oxidized with electron donation to the RS and lithium donation to the electrolyte solution. Electrons are shuttled from the negative to the positive electrode, resulting in an internal short circuit of the cell.

Agitation

Ojanen et al. (2018) showed that agitation of the solution was a promising option for industrial applications. However, the battery connector poles were rapidly corroded, leading to discharge and leakage of the internal battery components.

The discharge of the cells can be further accelerated by ultrasound cavitation effects or magnetic fields (Ali, et al., 2022). These methods increase the movement of the ions and therefore enhance the discharge of the cells. Yu et al. (2021) showed that the discharge rate can by increased by as much as 90% through subjecting the cells and discharge solution to ultrasound.

Cell vs module

Most commonly the module or the entire battery is immersed in the solution, as the intention of the stabilisation process is to make the battery safe to dismantle and extract the modules or the cells. However, the investigations of Xu et al. (2017) showed that the immersion of a battery pack results in higher risk of a high current electric arc forming, which could result in battery poles or shells fusing, resulting in the leakage of electrolyte, which could be ignited by the arc and burn on the water surface.

Industrial processes

There are several commercial/industrial processes that make use of chemical stabilisation techniques to prepare the batteries for recycling. This contradicts the claim of Amalia et al. (2024), who stated that there are "no reported commercial applications of battery discharging using the submersion method in an electrolyte solution". The details of the companies that make use of chemical stabilisation methods, together with a few details about the processes are given in Table 3.2. The names of companies in brackets are related technologies (previous names, subsidiaries, joint venture participants, etc.).

Table 3.2. Chemical stabilisation processes used in existing industrial LIB recycling.

Company	Notes
Bangpu Ni/Co High-tech Co.	Stabilisation of spent LIBs in brine solution.
Batrec Industrie	Stabilisation and storage of the batteries in water, followed by wet shredding u
BDT	Batteries are crushed under a sodium hydroxide solution without prior dischar
Cwenga Technologies	Shredding of batteries under aqueous solution of calcium chloride with gas rec
EcoBat (G&P Batteries, Promesa)	The Promesa process originally shredded the batteries under an aqueous solut
Euro Dieuze Industrie (SARP)	Stabilisation and storage of the batteries in water, followed by wet shredding u
Green Eco Manufacture (GEM)	Batteries are discharged by immersion in a sodium hydroxide solution before s
LiCycle	Submersion in a solution of $Ca(OH)_2$ and NaCl for discharging and shredding.
Neometals	Stabilisation using a brine solution (salt used is not disclosed).
Retriev (ToxCo)	Presently, the cells are crushed under an aqueous LiOH solution. This hydroly
SungEel HiTech (SungEel MCC Americas)	Stabilisation in a brine solution before shredding and hydrometallurgical proce
Taisen Recycling	Wet shredding of the batteries in water.

Discussion

The chemical stabilisation of LIBs is a cheap and easily implemented method. Due to them being cheaper than other salts, chloride-based salts, such as NaCl, are the most commonly used. The use of salt solutions for battery stabilisation has largely accepted without serious critical evaluation despite the risks that have been described (Liu, et al., 2019; Rouhi, et al., 2021). Recently, there have been critical evaluations of the use of salt solutions for the stabilisation of LIB cells by authors such as Ojanen et al. (2018), Shaw-Stewart et al. (2019) and Punt et al. (2022). Some of the biggest risks and concerns that have been noted regarding the use of chemical solutions for the stabilisation of batteries are the corrosion risks and the byproducts that could be produced through chemical stabilisation.

Corrosion risks

For the most part, the environmental impact of chemical discharging techniques has been neglected (Xiao, et al., 2020). Fang et al. (2022) observed that in almost all cases, good discharge capability of a solution was tied to corrosion being present. Segura-Bailón et al. (2024) also recognised that a higher corrosion level was linked to a faster discharge rate. The corrosion of a ferrous shell, when using a saline discharging solution, results in the risk of electrolyte leakage into the solution (Xiao, et al., 2020). This could result in a severe threat to the environment and human health if not managed correctly. The correlation could mean that the stabilisation is not being caused by chemical discharge, but rather by a physical process where the electrolyte is removed from the cells or else is contaminated to a point where a current can no longer flow. Li et al. (2016b) showed that the effluent wastewater from a battery stabilisation process using NaCl solutions contained high levels of aluminium, iron, and phosphorus as well as moderate levels of cobalt, lithium, copper, calcium, and manganese. It is probable that these chemicals have been leached out of the battery due to the corrosive nature of sodium chloride solutions. This means a loss of material, which could otherwise be recovered, as well as the need for an expensive effluent treatment facility. An additional hazard reported by Lee et al. was the generation of chlorine gas immediately upon immersion of the cells into a NaCl solution. The $LiPF_6$ from the electrolyte was found to leak from the battery due to the corrosion. According to Wang et al. (2022), the halide salts are the most corrosive, with carbonate and phosphate salts showing lower corrosion risks. Yao et al. (2020) captured the sediment from battery stabilisation processes using various discharge solutions and found that the mass of sediment in the NaCl solution was the highest, pointing to the NaCl solution having the highest degree of corrosion. The $MnSO_4$ solution produced no observable sediment as it has the lowest degree of corrosion. They also considered the concentration of metals in the solution after the discharging process and they found that the NaCl solution contained a significantly higher concentration of Li and Co than the other two solutions, meaning that there was likely a leakage of electrolyte and a leaching of the valuable Co. The corrosion risks destroy the ability to directly reuse materials from the cells and complicates the recycling process (Sommerville, et al., 2020). However, the cheap nature of chemical stabilisation methods mean that these methods are often still preferred despite the risks.

False stabilisation and rebound

Ojanen et al. (2018) showed that the battery terminals corroded “instantly within seconds, hindering the discharging reaction”. They went on to suggest that the reason that Lu et al. (2013) were able to achieve a complete discharge of a battery within 7 minutes in a 10 wt. % NaCl solution was not due to the electrochemical discharge of the battery, but in fact the loss of capacity of the battery due to disintegration of the components thereof. This raises a serious risk that many of the data reported by authors listed in Table 3.1 are inaccurate (Rouhi, et al., 2022). In almost all instances, halide salts have been shown to give the “fastest” discharging time, but this raises the question about whether this faster discharge time is primarily due to the movement of ions between the anode and the cathode within the cell, or due to corrosion of the cell resulting immobilisation of the ions within the cell. Ojanen et al. (2018) found that although it was impossible to bring a cell to a voltage of 0 V by solely using electrochemical discharging methods, it was possible to discharge a cell to below 2 V. Consequentially, their work suggests that all measurements that indicate a reduction of voltage to 0 V are likely erroneous due to corrosion occurring within the cells. In chemical stabilisation methods (section 3.4), the rebound appears to be independent of the discharge medium. Rouhi and co-workers (2021) investigated the possibility of using sequential discharge with multiple stabilisation and rebound cycles. They found that it was possible to lower the voltage output of the rebounded battery to below 2 V, but only after 4 discharge cycles over a period of almost 900 hours. They also investigated a long-term discharge of a battery (340 hours), and found that there was a reduced rebound, but that the rebound did occur. It is therefore important that the true voltage of the cell be measured to ensure sufficient stabilisation, as the voltage generated by a LIB cell can be decreased while undergoing discharge, and after rebound the voltage and SOC can be significantly higher (Rouhi, et al., 2021). One mechanism suggested to avoid the relaxation of the cells after discharging is to short-circuit the terminals once they have been stabilised (Diekmann, et al., 2017). Major concerns with chemical stabilisation are therefore, firstly, whether the perceived discharge is due to a reduction in the amount of lithium ions present within the cell, or a false discharge due to removal of the electrolyte through corrosion, and secondly, whether the reduced voltage achieved through chemical stabilisation will rebound when the cell is removed from the solution.

Emission of hazardous materials

Another major concern is the potential of chemical stabilisation methods to produce hazardous materials which could enter the environment. This is due to changes in the battery materials induced by the discharging process (Wang, et al., 2024). Cells are, after all, chemical reactors, and therefore, as they are pushed beyond their design limits, there are bound to be undesirable chemical reactions occurring, such as the dissolution of copper and subsequent deposition in different areas within the cells as Cu, Cu_2O or CuO (Wang, et al., 2024; Kim, et al., 2024). This results in the copper recovery being reduced. Two particularly undesirable cases are the generation of hazardous gases such as HF, and the leakage of per- and polyfluoroalkyl substances (PFAS) into the environment in the leach liquor.

Off-gases

Chemical stabilisation methods have a potential to generate HF particularly if there is contact between the electrolyte ($LiPF_6$) and water (Al-Thyabat, et al., 2013). The resultant gases from any reaction that occurs between these components include HF and POF_3 (Punt, et al., 2022; Diaz, et al., 2019). In addition, the $LiPF_6$ can decompose in dry environments to form LiF and PF_5. The PF_5 will also create HF and POF_3 if exposed to water. It is therefore important that the corrosion of the LIB does not result in the contact of the electrolyte with water.

PFAS compounds

In pyrometallurgical recycling processes for LIBs, the initial treatment steps are at temperatures that are sufficiently high to ensure that PFAS substances that are used in the LIBs are destroyed (Rensmo, et al., 2023). However, with chemical stabilisation processes that risk leaking the electrolyte from the cells, there is a risk of the emission of PFAS into the environment, because these are generally very stable at room temperature. This needs to be considered in the development of a cell discharging process using salt solutions, as corrosion of the cell container could lead to the contamination of the stabilisation solution with the electrolyte of other fluorinated liquids. In this case, it is important that the stabilisation solution is correctly treated, to prevent these PFAS from entering the environment.

Conclusions

A review was undertaken on the use of chemical stabilisation techniques that have been used in literature to pre-treat LIBs before they are recycled. The review of literature focused specifically on chemical stabilisation of the cells as a means to make the storage, transportation and processing of LIBs safer. It was found that a significant number of authors make use of chemical stabilisation techniques without fully understanding the risks associated with them. Most did not consider the impacts the stabilisation technique has on the downstream recycling processes and looked solely at what was the cheapest and fastest way to stop a cell from producing an electrical charge. Only a few studies highlighted the risks and problems associated with chemical stabilisation techniques. These include risks of corrosion and leakage of the electrolyte solution into the discharging solution, the generation of hazardous gases through the discharging process, and voltage rebound after removal from the discharge solution. Unfortunately, as these risks are, for the most part, not recognised by the users of the technique, it is likely that in many cases there is severe environmental impact that is overlooked. In the case of voltage rebound, users of chemical stabilisation techniques may be falsely led to believe that the batteries are discharged after submersion in a salt solution, and therefore that the batteries can be safely stored without the risk of fire or explosion. One way to avoid negative impacts of voltage rebound is shorting of the terminals immediately after stabilisation. An interesting observation from this study was that, although most literature focuses on the use of chemical stabilisation techniques, only a small portion of industrial processes use these techniques. Many industrial processes use in-situ chemical stabilisation techniques as they shred the batteries under either a salt solution (or water) to prevent fires and explosions.

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A. Appendix A

There are new LIB recycling processes and facilities being constructed and being brought online every month, and it is therefore impossible to produce an exhaustive list of stabilisation techniques used in existing industrial or commercial LIB recycling processes. However, Table A.1 gives a fairly comprehensive list of the processes that are made mention of in literature.

Table A.1. Extended list of stabilisation processes used in existing industrial LIB recycling.

Company	Notes
Accurec	No stabilisation of cells or smaller batteries, as first
AEA Technology	No stabilisation process, as the LIB are shredded in
AkkuSer	Manual sorting followed by crushing combined with
Anhua Taisen Recycling	*No information available*
Ascend Elements (Battery Resourcers)	Batteries are sorted based on SOH and energy recov
Aurubis[2]	*No information available*
Bangpu Ni/Co High-tech Co.	Stabilisation of spent LIBs in brine solution.
Batrec Industrie	Stabilisation and storage of the batteries in water, f
Battery Solutions[3]	“Room temperature, oxygen-free” mechanical proces
BDT[3]	Batteries are crushed under a sodium hydroxide solu
CATL (Brunpt)	No information on what technology is used in the di
Cwenga Technologies[2]	Shredding of batteries under aqueous calcium chlori
Dowa Eco-System	Dismantling of packs and modules followed by stabi
Dussenfeld	Energy recovery performed, but not full stabilisation
EcoBat (G&P Batteries, Promesa)	According to Pinegar et al., the batteries are stabilis
Ecopro	Operate a discharging process for battery packs, mo
Erasteel Recycling (Valdi)	Batteries are mixed with other feedstocks before fee
ERLOS	Stabilisation by energy recovery, followed by battery
Euro Dieuze Industrie (SARP)	Stabilisation and storage of the batteries in water, f
Farasis Energy[2]	Unspecified stabilisation process followed by remova
Fortum Oyj	Batteries are stabilised electrically with energy reco
GHTech	*No information available*
Glencore (Xstrata)	No stabilisation due to pyrometallurgical process. N
Green Eco Manufacture (GEM)	Batteries are discharged by immersion in a sodium
Green Li-ion	*No battery processing, as Green Li-ion only accepts*
GRS Batterien (Stiftung Gemeinsames Rücknahmesystem Batterien)	No stabilisation due to pyrometallurgical process. A
Guanghua Sci-Tech	Batteries are dismantled and shredded. There is no
Highpower International	Batteries are shredded before pyro- and then hydro-
Huayou Recycling	Battery packs and modules are automatically disma
Hydrovolt	Discharging of the batteries to “0V” using ‘manual p
Inmetco	The batteries are sorted, but there is no stabilisation
JX Nippon Mining & Metals Co. Ltd	Incineration of the cells removes the electrolyte and
Kobar	Batteries are stabilised, crushed, and screened befor
Kyoei Seiko	Batteries are not stabilised before co-processing, and
LiCycle	Submersion in a solution of $Ca(OH)_2$ and NaCl for
Lithion Recycling	Wet shredding in a light organic solvent with no pri
LithoRec (Chemetall Lithium, Albemarle Germany)	Energy recovery stabilisation method followed by to
Metal-Tech Ltd[3]	*No information available. Website refers only to tu*
Neometals	Stabilisation using a brine solution (salt used is not
Nickelhütte Aue	Thermal treatment of LIBs, with no mention of stab
Nippon Recycle Centre Corp.	*No information available*

Company	Notes
Northvolt AB (Revolt)	Energy recovery stabilisation method prior to disma
OnTo Technology	Cleaning and electrolyte removal by SC CO_2, giving
RecycLiCo Battery Materials (American Manganese Inc.)[2]	*No battery processing as only black mass or producta*
Redux	Energy recovery from batteries followed by thermal
Redwood Materials[1]	Batteries are fed directly into a rotating furnace, an
Retriev (ToxCo)	Originally used cryogenic freezing of the batteries in
SK tes (TES-AMM, Valibat Process, Recupyl)	No stabilisation, as preliminary shredding is underta
SNAM	Sorting of the batteries followed by pyrolysis to rem
Stena Recycling	Batteries undergo energy recovery stabilisation proc
Sumimoto (4R Energy Corp, Sony)	No stabilisation as the first process is calcination at
SungEel HiTech (SungEel MCC Americas[1])	Stabilisation in a brine solution before shredding an
Taisen Recycling	Wet shredding of the batteries in water.
Tele Battery Recycling (Telerecycle)	Mechanical processes used to shred the batteries (as
Tozero[2]	*No battery processing as only black mass or producta*
Umicore (VAL'EAS Process)	No stabilisation is required, as the initial process inv

[1] Proposed or conceptual processes

[2] Pilot plant and development stage

[3] Potentially no longer operational