



Review article



Immersion cooling for lithium-ion batteries – A review

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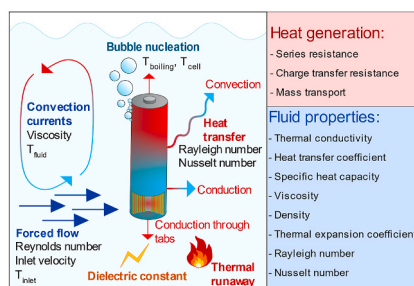
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HIGHLIGHTS

- Performance of battery immersion cooling and different cooling fluids reviewed.
- Immersion fluids can increase heat transfer by up to 10,000 times compared to air.
- Thermal properties of lithium-ion batteries and heat transfer mechanisms explored.
- Safety implications of battery immersion cooling discussed.
- Research gaps in battery immersion cooling presented.

GRAPHICAL ABSTRACT



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ABSTRACT

Battery thermal management systems are critical for high performance electric vehicles, where the ability to remove heat and homogenise temperature distributions in single cells and packs are key considerations. Immersion cooling, which submerges the battery in a dielectric fluid, has the potential of increasing the rate of heat transfer by 10,000 times relative to passive air cooling. In 2-phase systems, this performance increase is achieved through the latent heat of evaporation of the liquid-to-gas phase transition and the resulting turbulent 2-phase fluid flow. However, 2-phase systems require additional system complexity, and single-phase direct contact immersion cooling can still offer up to 1,000 times improvements in heat transfer over air cooled systems. Fluids which have been considered include: hydrofluoroethers, mineral oils, esters and water-glycol mixtures. This review therefore presents the current state-of-the-art in immersion cooling of lithium-ion batteries, discussing the performance implications of immersion cooling but also identifying gaps in the literature which include a lack of studies considering the lifetime, fluid stability, material compatibility, understanding around sustainability and use of immersion for battery safety. Insights from this review will therefore help researchers and developers, from academia and industry, towards creating higher power, safer and more durable electric vehicles.

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Nomenclature name			
ϵ	Dielectric constant	EV	Electric vehicle
κ	Thermal conductivity (W/mK)	FIA	Federation Internationale de l'Automobile
ρ	Density (g/L)	GTL	Gas-to-liquid
ν	Kinematic viscosity (cSt)	LIB	Lithium-ion battery
C_p	Specific heat capacity (J/kgK)	HEV	Hybrid electric vehicle
q	Heat flux (W/m ²)	HRR	Heat release rate
Acronym		LFP	Lithium iron phosphate
ARC	Accelerating rate calorimetry	NMC	Nickel manganese cobalt oxide
BN	Boron nitride	OCV	Open circuit voltage
BTMS	Battery thermal management system	PAO	Poly-alpha-olefins
CCC	Cell cooling coefficient	PCM	Phase change material
CEI	Cathode electrolyte interphase	PTC	Positive temperature coefficient
DEC	Diethyl carbonate	SEI	Solid electrolyte interphase
DMC	Dimethyl carbonate	SOC	State-of-charge
DSC	Differential scanning calorimetry	SOH	State-of-health
EC	Ethylene carbonate	SS	Silicone sealant
EMC	Ethyl methyl carbonate	THR	Total heat released
		TR	Thermal runaway
		UK	United Kingdom

1. Introduction

The deployment of lithium-ion batteries (LIBs) has rapidly increased with applications evolving from consumer electronics, to electric vehicles (EVs) and now to grid-scale balancing of renewable electricity generation. In these fields, uptake has been catalysed by the international push towards net-zero carbon emission targets with countries such as the UK committing to achieve this by 2050 [1]. However, improvements in: cost, energy density, power density, lifetime, safety, operating temperature, predictability and recyclability are still needed [2].

Effective battery thermal management systems (BTMSs) are now considered essential to maximise lifetime, enhance power capability and reduce the risk of thermal runaway. The aim of these systems is to remove heat from a battery pack, thus regulating the operating temperature, and to homogenise temperature within individual cells and between different cells of a pack. Many BTMSs currently exist ranging from passive air cooling to indirect liquid-based methods using cooling plates [3,4]. Liquid based systems are generally able to buffer and remove a larger amount of heat than air-cooled systems, due to their superior convective heat transfer coefficient and specific heat capacity. However, this additional performance often comes at the cost of increased complexity and system weight.

An emerging alternative to these BTMSs is the use of dielectric immersion cooling, whereby the cells are directly in contact with an electrically insulating working fluid. The advantage of this approach is that extremely high rates of heat transfer can be achieved through the direct contact of the cells with the immersion fluid, especially if 2-phase fluid systems are used. Here, the latent heat of vaporisation, associated with the liquid-to-gas transition, enhances the convective heat transfer, with effects such as nucleate boiling increasing the amount of turbulent mixing. Furthermore, many immersion fluids can act as fire suppressants, reducing the risk and effects of thermal runaway. However, to-date, immersion cooled systems have yet to be extensively implemented industrially at scale, due to challenges around cost of the fluids, uncertainty in system lifetime benefits and perceived weight penalties of integration. Some EV industrial applications, however, have started to emerge with notable examples including Kreisel who have adopted Shell thermal fluids [5], Xing Mobility which uses 3M's Novec fluid and Rimac Automobili who use Solvay's Galden fluids [6].

Therefore, this review paper presents the current state-of-the-art in BTMSs with a focus on the applicability, progress and challenges of

using immersion cooling for LIBs. Single-phase and 2-phase immersion fluids are discussed with emphasis on key metrics such as thermal conductivity, specific heat capacity and viscosity as well as their coupled nature with LIB performance which are highlighted in Fig. 1. Insights from this paper will therefore help LIB researchers and developers from academia and industry towards creating higher power, safer and more durable EVs.

2. Heat generation and thermal runaway of lithium-ion batteries

2.1. Coupled electrochemical and thermal behaviour

The performance of a battery is highly thermally coupled [7] and therefore understanding of the thermal properties of a cell, its heat generation characteristics and resulting electrochemical behaviour is important. In terms of the coupled thermal and electrochemical behaviour, this can be broadly divided into thermodynamic and kinetic aspects. For thermodynamic factors, the temperature affects the open circuit voltage (OCV) of a battery. However, the magnitude of this is relatively small, with Troxler et al. [8] showing that showing the dV/dT response of a Nickel Manganese Cobalt oxide (NMC)/graphite cell only varies between -5×10^{-4} and 1×10^{-4} V/K. The more critical thermally coupled aspect is the kinetics, whereby factors such as the conductivity and charge transfer resistance of a battery are highly thermally coupled. Aspects of heat generation relevant to immersion cooling will be expanded here but readers are referred to other review papers such as Bandhauer et al. [9] for a comprehensive review of thermal issues in LIBs.

Temperature distribution within cells is evidently an important parameter in determining a LIB's longevity. Two key thermal parameters are the specific heat capacity, C_p (J/kg. K), and the thermal conductivity, k (W/m.K) which describe the heat stored and the heat flow rate, respectively. In the case of LIBs, their construct consists of alternating layers of materials which leads to anisotropic thermal properties.

For thermal conductivity, the in-plane value is often an order of magnitude higher than the through-plane value [10]. Yet, the exact values are dependent on cell design and materials used. Wei et al. [11] reviewed the thermal conductivity of various battery form factors and chemistries, and quoted ranges of approximately 10–45 W/m.K for in-plane/axial conductivity, compared to 0.6–1.5 W/m.K for through-plane/radial conductivity for pouch and cylindrical cells,

respectively. In practice, the interaction between the layers has been shown to be more complex, with thermal contact resistances between layers having a large impact on the total thermal conductivity [12]. Furthermore, the electrolyte has been shown to significantly reduce this contact resistance, with the measured thermal conductivity of a pouch cell varying by as much as 92% with and without the electrolyte present [10]. Due to the complexities in modelling the thermal contact resistances of porous materials, with this being a function of the amount of electrolyte and the applied pressure, the most reliable way to determine thermal conductivity remains to experimentally measure it. Steinhart et al. [13], for instance, measured the thermal conductivity of a prismatic cell under different levels of compression. Here, they noted that when increasing the externally applied pressure from 37.1 kPa to 74.2 kPa (manufacturers recommended limits) the through-plane thermal conductivity increased by 11.9%, which they attribute to the lower thermal contact resistance.

Furthermore, both specific heat capacity and thermal conductivity are often assumed to be constant, however studies have shown that these thermal properties can vary over their range of use. Zhang et al. [14], for instance, quoted various single specific heat capacity values of different battery chemistries ranging from 896 to 1720 J/kg.K, whereas Bazinski et al. [10], showed that, for a lithium-iron phosphate (LFP) cell, the specific heat capacity varied by over 10% between 15 °C and 35 °C. Therefore, accurate thermal modelling of a battery should also consider parameter variation over the range of use.

2.2. Heat generation/dissipation and thermal gradients

LIBs can generate or absorb heat through irreversible and reversible processes, with the irreversible terms usually dominant, resulting in a net positive heat generation. The processes behind this are: electronic resistance, electrochemical reactions, phase changes and diffusion [15]. The heat from diffusion, can be further broken down into the heat from diffusion within particles and across the electrolyte, with the heat from diffusion across the electrolyte often quoted as smaller [16]. Often these sources of heat are a function of state-of-charge (SOC), temperature and current [17], with these relationships changing with cell age.

Almost all large battery packs now feature an active cooling system, both for increased safety and for increased battery lifetime. In an active cooling system, the heat is extracted using a coolant causing tempera-

ture gradients to build up within the cell and throughout the cooling system. To illustrate these temperature gradients, Fig. 2 considers 1D steady state heat transfer. This is a good approximation of the through thickness temperature gradient in a large format pouch cell when surfaced cooled on both sides of the cell. Applying the 1D steady state assumptions with boundary conditions to the heat diffusion equation yields Equation (1). Where ΔT_{Cell} is the temperature gradient in the cell, T_{Max} is the maximum temperature, $T_{Surface}$ is the surface temperature, $\dot{e}_{gen}$ is the heat generated per unit volume and L is the characteristic length. This shows that the steady state thermal gradient within a cell is governed by the geometry and thermal conductivity, for a given heat generation. The relationship between the coolant temperature and the cell surface is given by Equation (2). Where ΔT_{Fluid} is the temperature gradient between the cell surface and the fluid temperature, where T_{∞} is the fluid temperature and h is the convective heat transfer coefficient. Fig. 2, then illustrates the magnitude of these temperature gradients for different heat generation rates and convective heat transfer coefficients. Note the large influence that the convective heat transfer coefficient has on ΔT_{Fluid} .

$$\Delta T_{Cell} = T_{Max} - T_{Surface} = \frac{\dot{e}_{gen} L^2}{2k} \quad \text{Equation 1}$$

$$\Delta T_{Fluid} = T_{Surface} - T_{\infty} = \frac{\dot{e}_{gen}}{2h} \quad \text{Equation 2}$$

Internal resistance has been shown to be highly sensitive to changes in temperature which, in the case of a cell comprised of many layers, results in imbalanced resistances in parallel cell layers, leading to non-uniform current flow and uneven localised SOC. This uneven current flow will cause heterogeneous degradation and has been shown to reduce the lifetime of a cell by 65% [18]. Tranter et al. [19], investigated this uneven current distribution in a cylindrical 18650 cell which demonstrated the magnitude of this current heterogeneity increasing with increasing C-rate, where C-rate is defined as the ratio of the current (A) to the nominal capacity (Ah) of the cell. In their work, they also showed how this can, to a certain extent, be homogenised by increasing the number of tabs on the electrode. Ideally, cell and pack gradients would be minimised by fixing every surface of the cell, including the tabs, at the same temperature. From a heat transfer perspective, the ideal coolant would therefore have an infinitely high convective heat

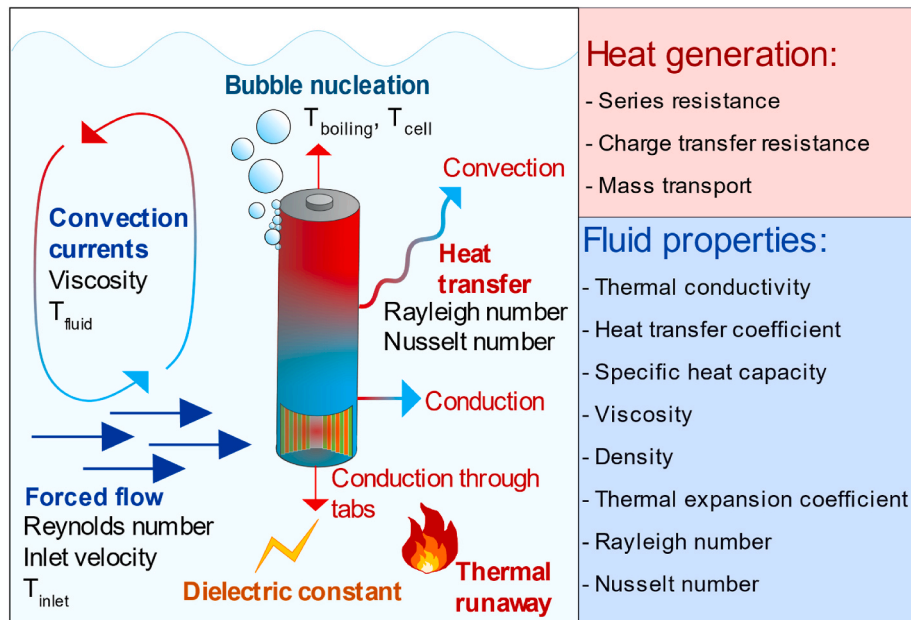


Fig. 1. Overview of various considerations in immersion cooled battery thermal management systems.

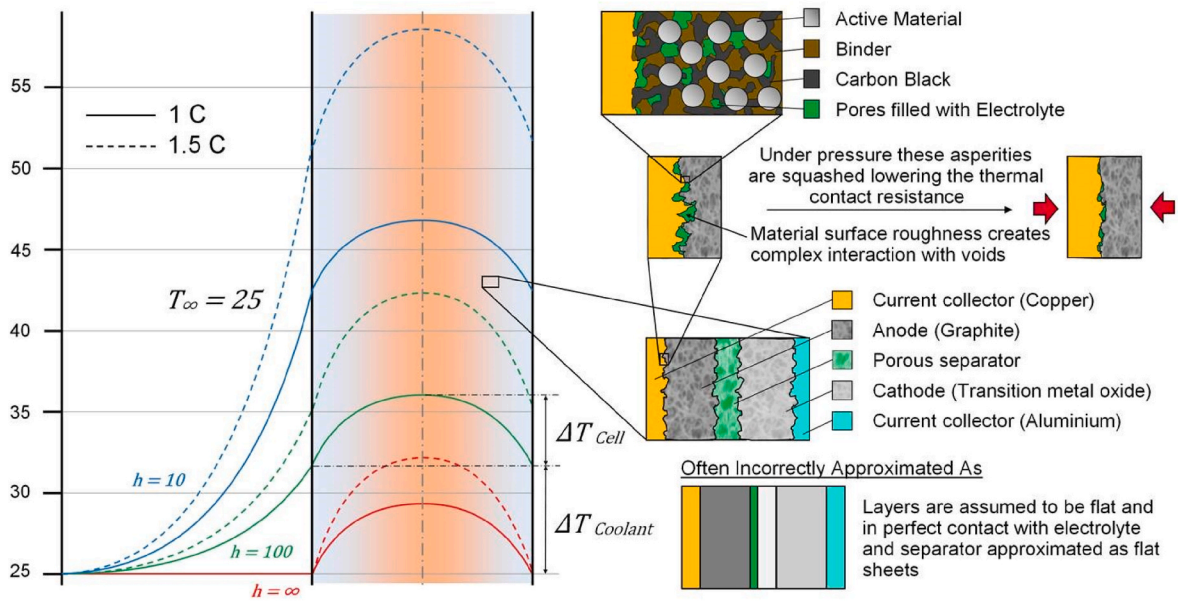


Fig. 2. Typical 1D temperature profiles in the coolant fluid and through the thickness of a cell.

transfer coefficient, as shown in Fig. 2, and also be in direct contact with every surface of the cell. This is where immersion systems using dielectric fluids offer huge potential as they allow the whole cell, including the tabs and the bus bar to be immersed in the coolant.

2.3. Degradation effects

Degradation of LIBs involves complicated multi-physical mechanisms including many thermal, electrochemical and mechanical phenomenon, which have been extensively studied [20–23]. In this review, the focus is mainly on systems to negate temperature related degradation processes, with Fig. 3 highlighting the key factors affecting temperature related degradation.

Chen et al. [24] suggested that the optimal temperature range for a LIB is between 15°C and 35 °C. Outside of this temperature window, unwanted side reactions, which are highly temperature dependent, occur leading to accelerated degradation of the cell. Determining the optimal operating temperature requires a balanced view between performance and lifetime. Material properties including electrolyte conductivity, diffusion coefficients in the electrolyte and electrode particles

are generally more favourable at high temperatures [25], with both the reaction kinetics [26] and material properties [25] following the Arrhenius equation. However, this also enhances the unwanted side reactions. For example, growth of the solid-electrolyte interphase (SEI) on the anode, growth of the cathode electrolyte interphase (CEI) will be accelerated, consuming more electrolyte and available lithium-ions, leading to increased impedance and heat generation [20,21]. Kumai et al. [27] Identified that gas formation may occur due to electrolyte decomposition, creating bubbles, blocking ion transportation, and possibly leading to exfoliation of graphite layers [21]. This electrolyte decomposition, may well also negatively impact the thermal properties of the cell, mentioned previously, however few studies have investigated the evolution of thermal properties with aging.

A linear relationship between the logarithm of capacity loss and the inverse of temperature, which is consistent with the Arrhenius equation, was observed during calendar degradation tests [26,28]. Low temperature operation will also have negative influence on the battery's lifetime as lithium plating becomes the dominate side reaction, resulting in severe loss of available lithium-ions [29]. This highly active plated lithium metal could then react with the electrolyte once the temperature

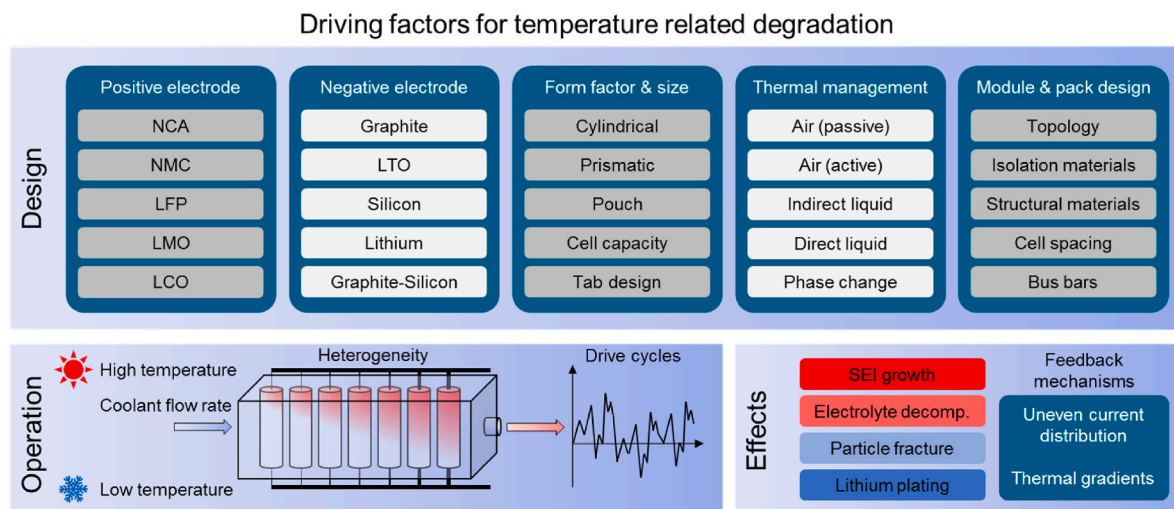


Fig. 3. Factors influencing different thermally coupled degradation modes.

increases [30]. Therefore, it is important to control the battery temperature within the optimal range to ensure prolonged lifetime.

The temperature heterogeneity across the battery is another important factor determining the battery life. Temperature heterogeneity is affected by the design, manufacture and operation of cells, as indicated in Fig. 3. Design factors, such as the location, numbers and size of tabs play an important role on the temperature distribution [31,32]. Arunachala et al. [33] identified that this is due to the ohmic heating contribution of the tabs, where the electron flow is focused during cycling. Therefore, heat generation near the tabs is often more pronounced. Lee et al. [34] investigated the optimal tab position on pouch cells and found that batteries with tabs on the same side exhibit larger temperatures and stress gradients than those with tabs on the opposite side. Large tabs are beneficial for heat dissipation especially for the tab cooling strategy proposed by Hunt et al. [18]. In addition, Zhao et al. [31] proposed that increasing the number of tabs helps to inhibit thermal gradients by reducing the in-plane electron transport losses through long electrodes. Other factors which should be considered for cell degradation are non-uniform stresses which occur at the curved part of the jelly roll of prismatic cells [35] and the centre of cylindrical batteries [36]. Osswald et al. [36] discovered that the inner most part of the cell had higher low-frequency impedance which was caused by the increased pressure of the cell winding at the centre of a cylindrical cell. When considering all these factors it is evident that heterogeneous cell performance is impacted by many factors.

A more uniform temperature distribution, whilst being achievable through design and manufacture of the cell, still amplifies heterogeneous performance and accelerates degradation so homogenous temperature distribution through the use of an external BTMS is essential for the longevity and performance of LIBs.

2.4. Battery packs

Battery packs are comprised of many series and parallel connected cells to achieve a practical voltage and capacity. The interaction between cells in a pack is complex mainly due to; variations in the cells such as their SOC, state-of-health (SOH) and manufacturing variance [37,38], additional contact resistances which can vary between 0.05 and 0.35 m Ω depending on the joint type, pressure and contact area [39], and also the thermal gradients which can exist across a battery pack. Localised temperatures in the pack result in very uneven current flow and heat generation [40,41]. All these factors combine dynamically resulting in what can be a highly unbalanced resistor network.

In the short-term, the available power and the useable capacity can be limited as a direct result of pack thermal gradients, as the most worked cells hit their voltage safety limits earlier [9,41]. Additionally, the non-uniform current flow increases power losses as the loss varies with current squared. Long term effects come in the form of increased cell degradation which reduces pack lifetime [42]. Larger currents lead to higher rates of non-linear degradation and the heterogeneous current flow puts significant extra strain on the cells that take a higher than average current which results in increased internal resistance and a reduction in useable capacity [43]. Studies have investigated this current heterogeneity within pouch cells and also within small packs [38]. Therefore, it is clear that, in the presence of thermal gradients, degradation is increased, though improved coolant designs this may extend the lifetime of a pack, with authors such as Darcovich et al. [44] suggesting improvements from 3.5 to 8 years.

2.5. Thermal runaway

Avoiding thermal runaway (TR) is one of the key criteria for BTMSs. The root cause of TR may be induced by mechanical, electrical and/or thermal abuse [45–47]. Mechanical abuse is generally caused by mechanical deformation of the battery, which occurs during vehicle collision, and the subsequent extrusion and puncture, leading to partial

rupture of the separator and can short the cell. Electrical abuse is generally caused by external short circuits, overcharge or over discharge of the battery, which leads to dendrite growth and penetration of the separator. Thermal abuse is generally caused by the inconsistencies between the internal resistance and heat dissipation of the single cell in the pack, and abnormal increase of the contact resistance, which leads to the large-scale collapse of the separator [48]. Here, internal short circuit is a common link between the processes to initiate TR [49]. Mechanical abuse leads to cell deformation and the potential formation of an internal short circuit, leading to electrical abuse. This electrical abuse is then accompanied by joule heating and chemical reaction heat, which leads to thermal abuse. Subsequently, thermal abuse leads to the rise of temperature, which in turn triggers the TR chain reaction. TR in a single cell can propagate through convection, radiation and conduction, to other cells, and then leads to TR of the entire pack [46].

The heat release rate (HRR) is an important parameter to evaluate the fire risk of LIBs, with the total heat released (THR) obtained by integrating the HRR curves. The size of the HRR and THR depends on the specific energy, capacity and SOC of the battery. Based on the HRR and THR, batteries with 100% SOC have the highest THR. Studies have found that the THR of a 18650 LFP battery at 100% SOC was 150 kJ, and the THR by a 18650 LCO battery was 230 kJ [50,51], highlighting the chemistry sensitivity to the risks associated with TR. As cell form factors increase, so does the THR, as exemplified by a 68 Ah LFP battery which contained 6600 kJ of energy. As an absolute metric, the HRR of LIBs at 100% SOC state is almost the same as that of petrol, and at 50% SOC is the same as that of the combustion of polymers such as polymethyl methacrylate. In addition, the external environmental pressure is directly proportional to the HRR and THR, the lower the environmental pressure, the lower the HRR and THR of LIB [52]. This is mainly because with the decrease of pressure, the absolute concentration of oxygen at the surface of the battery decreases, which leads to the reduction of combustion reaction rate and the reduction of flame thermal feedback. In addition, low pressure, leads to the weakening of the thermal convection effect of flame on the lithium battery, and also affects the HRR and THR. Assessing whether a BTMS can prevent TR depends on the SOC and chemistry of the battery. Feng et al. [46] presented a comprehensive review of these mechanisms as well as comparison of the HRR of different components in a cell, highlighted in Fig. 4. Here, accelerating rate calorimetry (ARC) and differential scanning calorimetry (DSC) tests, from various literature sources, were compiled to create this overview, where the chemical kinetics correspond to a cell at 100% SOC [46].

It should also be noted that during thermal runaway venting occurs. Venting typically accompanies TR of LIBs due to the extremely high internal pressures generated [53,54]. During the process of venting, various materials are ejected from the cells, including gases, liquids and solids [55]. Gaseous ejection products mainly contain carbon dioxide (CO₂), carbon monoxide (CO), hydrogen (H₂) and short-chain hydrocarbons, such as methane (CH₄) and ethylene (C₂H₄), most of which are flammable [56,57]. The composition, concentration and volume of gaseous ejection products vary with many parameters of the cell, including: SOC, cathode material and electrolyte [58,59]. Besides this, Golubkov et al. [59] verified the existence of fluoride gases such as hydrogen fluoride (HF) and phosphoryl fluoride (POF₃). Although the amount may be small, they can be toxic. As for liquid ejection products, Zhang et al. [57] detected water and electrolytes including ethyl methyl carbonate (EMC), diethyl carbonate (DEC), dimethyl carbonate (DMC) and ethylene carbonate (EC). These organic vapours can threaten human health. A summary of the various gaseous and liquid ejection products, detected with a combination of gas chromatography, mass spectrometry and ion chromatography [55], are listed Table 1. Additionally, solid ejection products can also be dangerous due to their elevated temperature, toxicity and environmental pollution. The elemental compositions contain carbon (C), nickel (Ni), copper (Cu), cobalt (Co), manganese (Mn), aluminium (Al) and lithium (Li) in order of content from the high

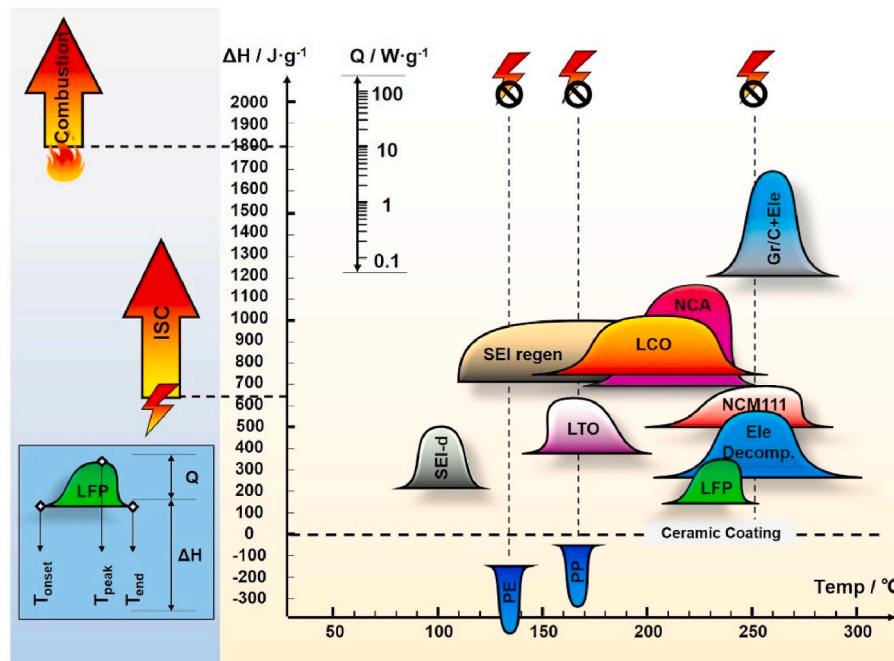


Fig. 4. Energy release diagram showing the thermal metrics in different components of the battery [46].

to the low [57,60,61]. Awareness of the various materials which are ejected from a cell is therefore crucial to develop robust BTMSs, especially in the case of the flammable components, which may necessitate non-flammable cooling fluids.

3. Battery thermal management systems

The main types of BTMS include air cooling, indirect liquid cooling, direct liquid immersion cooling, tab cooling and phase change materials. These are illustrated in Fig. 5 and in this review, the main characteristics of non-immersion cooled systems are briefly presented, with insights and key metrics presented towards providing context for a deeper discussion around immersion cooled systems in Section 4.

A list of key metrics for typical BTMS working fluids is shown in Table 2. These liquid cooled systems can be subdivided based on the means by which they make contact with the cells, which includes: (a) indirect cooling where coolant is isolated from batteries via a jacket, tube or plate adjacent to battery modules; (b) direct cooling (immersion cooling) where batteries are directly in contact with the coolant.

3.1. Air cooling

Air cooling systems are one of the most widely used BTMSs in EVs due to low cost, simple design, low weight, easy maintenance, and no leakage issues compared with other cooling systems [79–81]. In this system, air removes heat via constant flow between the batteries.

Table 1

Composition of ejections during thermal runaway adapted from [55,56,60].

	Gas ^a	Liquid	Solid ^b
Level 1	CO ₂ , CO, H ₂	Electrolyte (DMC, EMC, DEC, EC), H ₂ O	C, Ni, Cu, Co, Mn, Al, Li
Level 2	CH ₄ , C ₂ H ₄ , C ₂ H ₆ , C ₃ H ₆		
Level 3	C ₃ H ₈ , C ₄ H ₁₀ , C ₄ H ₈ , C ₂ H ₂ , C ₃ H ₄ , C ₄ H ₆ , C ₅ H ₁₀ , HF, HCl	C ₅ H ₁₀ , C ₆ H ₁₂ , C ₆ H ₆ , C ₅ H ₁₂ , C ₉ H ₁₈	K, P, Fe, Na, Ca, Sr, Cr, Ti, Zr, B, Mo, Zn, Mg, Sn, Ba

^a Under 25 $^{\circ}\text{C}$ and 101 kPa.

^b List of ejected elements.

Estimates suggest that air cooling could handle normal EV driving conditions if heat rejected from the battery pack is lower than approximately 4 kW [82]. Generally, air cooling systems can be divided into two main categories: active air cooling (forced convection) and passive air cooling (natural convection) [83,84]. The active air cooling system possesses higher cooling effectiveness as the air is forced to flow into the battery pack enhancing the convective heat transfer, however this requires additional parasitic energy from fans, with additional weight and volume associated with fan ducts and manifolds [85,86]. On the contrary, there is no parasitic power consumption for passive air cooling systems with only natural air flow which results in inferior cooling performance [87]. Therefore, active air cooling system accounts for a larger market proportion and has been adopted for several cases such as cooling systems of the Nissan e-NV, Toyota Prius, and Honda Insight.

Improvements to the effectiveness of air cooling systems have been made in recent years. This centres around three main aspects: cell arrangement, air flow path, and flow rate. For example, Fan et al. [88] conducted a 3D transient thermal analysis of a prismatic cell module with a forced air cooling system where they lowered the spacing between cells and increased the air flow rate which reduced the maximum temperature. Yet, despite this progress, air cooling systems are insufficient for more aggressive operation. The root cause is that air has lower heat capacity as well as lower thermal conductivity compared with other mediums (e.g. liquids and phase change materials); reducing cooling capabilities and causing poor temperature uniformity in the battery pack [89,90]. Hence, air cooling might not be appropriate for next generation EV with larger size battery packs requiring ultrafast charging. Nevertheless, air cooling systems still have a promising prospect in future BTMSs requiring only low thermal dissipation [24].

3.2. Liquid cooling

Liquid cooling encompasses both indirect liquid cooling and immersion cooling. Given the limitations of air cooling systems, liquid cooling is an alternative route for large scale EV BTMSs [91]. Compared with air, liquids have higher specific heat capacity as well as better thermal conductivity [92].

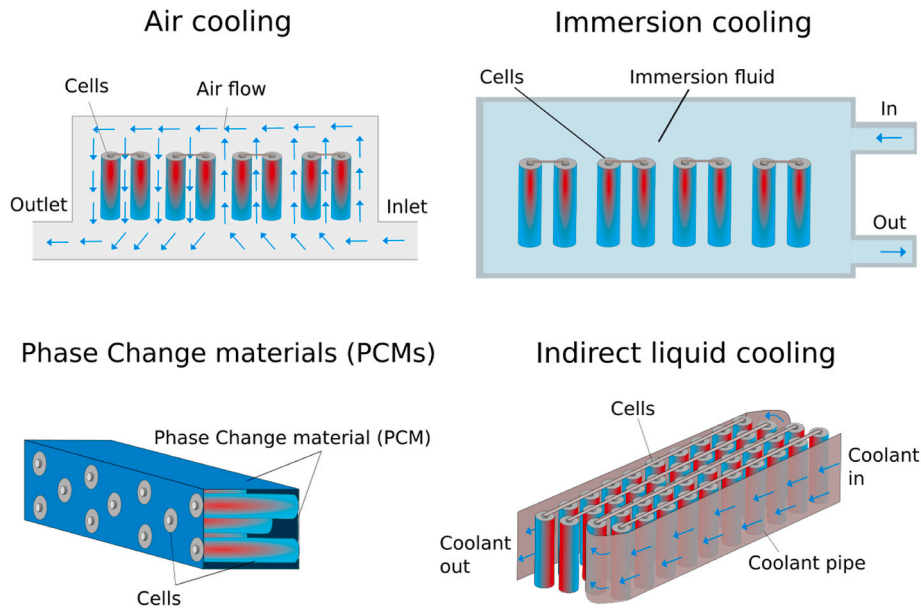


Fig. 5. Different thermal management systems adapted from [62].

Table 2

Comparison of various thermal management systems for EVs.

BTMS	Thermal conductivity of coolant (W/m.K)	Specific heat capacity (J/kg.K)	Convective heat transfer coefficient W/(m ² .K)	Dielectric constant
Air cooling	0.0242 [63]	1,006 [63]	10–100 [64]	1.0 [65]
Indirect liquid cooling (water-glycol)	0.3892 [63]	3,323 [63]	10,000 [66]	64.9 [67]
Immersion cooling single phase	0.129–0.15 [68,69]	1,370–2,241 [69,70]	2,000 [71]	2.1–2.8 [69,72]
Immersion cooling two phase	0.075 [73]	1300 [74]	20,800 [71]	7.4 [74]
Tab cooling	8,225–9,611 [75]	-	3,000 [75]	-
Phase change material	0.15–0.40 [76]	1770 [77]		6.3–13.3 [78]

3.2.1. Indirect cooling system

Indirect cooling is one of the most widely used EV BTMS due to its ability to maintain a good uniform pack temperature distribution, favourable specific heat capacity, and good thermal control [93,94]. Mixtures of water and ethylene glycol are common coolants. Normally, the coolant flows along channels of pipes or cooling plates, carrying the rejected heat out of the battery pack [95,96]. Based on the cooling channels' position relative to the batteries, indirect cooling systems can be divided into bottom cooling [97] and side cooling [98]. The Tesla Model 3, for instance, uses the side cooling method with serpentine tubing attached to cylindrical cells [99]. The Audi e-tron, Porsche Taycan, and Mercedes-Benz EQC on the other hand install the cooling plate underneath the cells with the bottom cooling method.

Tremendous efforts have been made in the past few years on the design of the cooling channel structure and improving the thermal properties of the working fluids, the latter will be discussed in Section 4. Darcovich et al. [100], for instance, compared the thermal performance between 2 indirect cooling systems: side cooling and bottom cooling for prismatic cells, through coupled electrochemical and thermal

simulations. The side plate system enabled a lower maximum temperature and better temperature uniformity than the bottom cooling plate system due to the more comprehensive contact with the battery.

Recently, Li et al. [101] presented the thermal response of a prismatic battery with a liquid mini-channel cooling plate under 5C fast charging and external shorting conditions. With adequate coolant flow velocity, the maximum cell temperature and temperature gradient were limited to below 30 °C and 5 °C, respectively. For external shorting conditions, with a 0.1 m s⁻¹ coolant flow velocity, the cell could be controlled to below 60 °C; preventing thermal runaway. Based on the thermal performance for a single cell, the author also studied a 50 V battery pack comprising 15 prismatic cells. The maximum cell temperature and maximum temperature difference in the pack were controlled to be below 40 °C and 6 °C under a 5C discharge.

Heat pipes are another form of indirect liquid cooling. These offer extremely high thermal conductivities through the evaporation and condensation of a 2-phase working fluid contained within a thermally conductive sealed pipe. Reviews such as Jouhara et al. [102] provide an overview of their broad application with reviews such as Kim et al. [96] focusing on their performance and challenges in BTMSs. Here working fluids such as water and acetone are common, however, whilst superior heat transfer can be achieved over pure metallic heat sinks, making effective thermal contact was identified as a key challenge. Furthermore, many studies suggest that forced air cooling would be needed to condense the working fluid and ensure suitable temperature operation due to the limited heat capacity of the heat pipes. Behi et al. [103], for instance, modelled the performance of a heat pipe cooling system in a high-power prismatic lithium titanate battery pack under 8C discharge. Here they calculated an effective thermal conductivity of 8212 W/m.K but noted that a single heat pipe only provided 29.1% of the required cooling load and that thermal gradients in large cells can impact the performance of the heat pipe. Thus, various hybrid systems have been proposed which combine the high specific heat of PCMs and the high thermal conductivity of heat pipes [104]. For instance, Huang et al. [105] investigated a hybrid system for a 30 cell 18650 pack which used a PCM of paraffin with graphite and a heat pipe which was cooled either by air or liquid. Here they noted that for a pure PCM system, the temperature difference would reach ~3.5 °C, with this increasing with cycles due to thermal accumulation, as opposed to <1 °C for their combined system.

Although indirect cooling systems have received considerable focus,

some potential drawbacks still need to be solved. Compared with air cooling systems, the complexity of the whole system is much higher due to the additional components (e.g. chiller and radiator) and complicated piping. This results in more potential sources of failure, additional weight and coolant leakage issues. Beyond this, the thermal resistance between the cell and coolant is often increased due to the existence of cooling tubes/plates as well as imperfect thermal contact, which might be an application barrier to ultrafast charging conditions (300 kW) [106].

3.2.2. Tab cooling

Recent progress on pouch cell tab cooling has been proven as a promising way to achieve high temperature uniformity. For instance, Hunt et al. [18] compared the electrochemical performance between surface cooling and tab cooling with a 5 Ah pouch cell with tabs on opposite ends of the cell. The two cooling methods exhibited similar capacity loss under lower current operation of C/20 but at the higher rate of 6C this accessible capacity loss was more apparent. The loss of useable capacity for the pouch cell with surface cooling was 9.2%, while the pouch cell with tab cooling demonstrated a stable performance with only 1.2% loss of useable capacity. After 1000 cycles, surface cooling caused a capacity loss which was three times higher than cell tab cooling. The enhanced electrochemical performance of tab cooling was attributed to more homogeneous temperature distribution, leading to more homogeneous current distributions between cell layers.

Subsequently, the same group developed a 2D electro-chemical model to observe the effect of thermal gradients on cell performance [107]. Here, Zhao et al. [107] showed that, for the same 5 Ah cell (6C discharge), surface cooling delivered a lower average temperature but larger thermal gradient than tab cooling despite a notably large thermal resistance at the cell tab. However, cell tab cooling is not always better than surface cooling. For instance, Dondelewski et al. [108] showed that in the case of a large form factor 20 Ah LFP pouch cell where the tabs were on the same side, pure tab cooling was not able to remove sufficient heat, resulting in a higher rate of degradation compared to surface cooling. Zhao et al. [109] further explored this thought modelling and testing different cell tab configurations and showed that by increasing the tab width from 30 mm to 70 mm the peak cell temperature could be reduced by 14%. In the case of increasing the width of the tab, surface cooling was still superior in heat removal capabilities, however they noted that with increases in the thickness of the tabs, the performance of tab cooling could be comparable with surface cooling. Alternative designs include the “tab-less” 4680 cylindrical design proposed by Tesla and Panasonic, which was modelled by Tranter et al. [110]. Based on their results, the “tab-less” design could reduce ohmic losses by as much as 5 times compared to traditional tabbed designs. Yet, the exact impact depends on the heat generation and design of the cell.

To address this issue, Hales et al. [111] introduced the surface Cell Cooling Coefficient (CCC), which is a tool to help designers select the proper BTMS as well as assess pack-wide thermal gradients. The CCC is a BTMS specific measurement for the cells rate of rejecting heat at a given temperature gradient. This allows for a comparison to be made between BTMS as well as cell size and capacity.

Considering most use cases in real-world application are still dominated by the surface cooling method, tab cooling may be promising due to its ability to impose favourable temperature gradients, and potential to extend cell lifespan. Nevertheless, two bottlenecks still needed to be taken into account before tab cooling can be upscaled. Firstly, tab cooling may not be as effective in controlling the maximum cell temperature, especially for harsh utilization conditions. Secondly, the practicalities of tab cooling systems designs are still being debated, with the ease of system manufacturability a key issue. The tab should be electrically isolating but most cooling tubes are aluminium. How to accelerate the heat transfer rate while maintaining the tabs in an insulation environment is still, therefore, under debate [112]. Immersion may provide a more simplistic version of tab cooling as the tabs can be

inadvertently cooled while the cell is immersed without the need of a complex tab cooling system.

3.3. Phase change materials

Phase change materials (PCMs) have also received significant attention in BTMSs [113]. The latent heat associated with the material, as it undergoes a phase transition during heating, affords it with favourable specific heat capacity characteristics. This can come in different forms with the main classifications including: solid-liquid (covered in this section) and liquid-gas (covered in Section 4) PCMs. When selecting a PCM for a BTMS, the melting point, specific heat capacity and thermal conductivity are often key figures of merit [114].

One of the main organic PCMs used is paraffin, with the length and formation of carbon within the chains giving the paraffin different physical and chemical properties. The melting point of paraffin is dependent on the length of the straight chain alkanes and the mixture combination of alkane chains. The specific heat capacity of paraffin wax ranges from 2140 to 2900 J/kg.K [115] which is higher than many immersion coolants.

Many PCMs generally have poor thermal conductivities leading to poor thermal propagation and therefore low utilization of the potential specific heat capacity in the system. This challenge has motivated many researchers to add different additives to PCMs to tune the thermal properties. These include various additives such as: carbon fibres [116], nano-magnetites [76], metal foams [117–120] and expanded graphene [121]. The improvements in thermal conductivity in many cases can be significant. Şahan et al. [76], for instance, investigated paraffin-nano-magnetite (Fe₃O₄) composites for use in thermal storage. Using diffusivity and conductivity measurements, they showed that paraffin with the nano composite increases thermal conductivity of the paraffin by 48% with 10 wt% of the nano-magnetite without changing the melting point of the base paraffin. Although thermal conductivity is an important factor which needs to be considered when choosing the additive and the proportions used; uniformity of heat dissipation and homogeneity of the PCM with additive is also important. Motivated by this, PCM composites can also be used in conjunction with other forms of thermal management systems such as heat pipes in order to improve the temperature uniformity during heat transfer from cells to the heat pipe [122]. Yet, despite the various advantages of PCMs, direct cooling of the cell remains a challenge due to the thermally conductive additives also reducing the dielectric properties of the material and solid-phase heat transfer being limited to conduction.

4. Immersion cooling

As one of the emerging cooling technologies in recent years, direct liquid cooling, which is also called immersion cooling, has attracted considerable attention for electronic devices and in the EV industry [98, 123–126]. In this system, the battery is submerged into a non-conductive dielectric fluid, thus making direct contact with the cell. Candidate dielectric fluids have included: hydrocarbon oils, silicone oils and fluorinated hydrocarbons. This unique way of cooling brings several advantages. Firstly, immersion cooling has the potential to provide the best pack and cell temperature uniformity among all the cooling methods. This is because all battery surfaces are in the fluid, providing a homogeneous, high heat capacity thermal transport path for heat rejection. This direct contact with the cell surfaces further reduces the thermal contact resistances experienced in indirect cooling systems [98]. Immersion cooling simplifies the system design and reduces the system complexity [123]. Furthermore, the suppression of thermal runaway is usually observed for immersion cooling systems as some of the dielectric fluids are also flame-retardants, enhancing the safety of the LIB pack. Different embodiments of immersion cool depending on the degree of submersion, type of flow and regime of fluid operation exist which are highlighted in Fig. 6.

Recent research in immersion cooling has mainly centred around performance analysis for single cells and battery packs. Nelson et al. [127] thermally modelled a 48-cell system with both direct cooling and air cooling. The result showed that direct cooling with silicone oil exhibited superior heat dissipation with the cell temperature rise only 2.5°C , compared to air cooling which exhibited a 5.3°C under the same load conditions. The similar conclusion on thermal performance comparison between direct silicone oil cooling and air cooling was also shown by Karimi et al. [69,128]. In other works, Kim and Pesaran [63] conducted a systematic single cell simulation comparison for three different thermal management systems including air cooling, indirect water/glycol jacket cooling, and direct mineral oil cooling. Among the three cooling methods, direct cooling demonstrated the lowest maximum cell surface temperature difference and the best heat transfer coefficient, especially with smaller cooling channel diameters and higher flow rates. Beyond water/glycol systems, authors have also proposed working fluids such as a pressurized saturated liquid ammonia. Here, Al-Zareer et al. [129] showed that a pressure of 9.0 bar with this liquid only covering 5% of the surface of the cell is adequate to maintain the battery temperature below 40°C for high power charging and discharging cycles at a rate of 7.5C.

Of course, total system effectiveness needs to be considered for any BTMS. Park et al. [130] modelled cylindrical cells for immersion cooling and found that a narrow battery module with small gaps consumes less parasitic power than a wide module with large gap due to the low coolant flow rates, while the cell-to-cell temperature variations for a wide module with large gaps could be diminished. They found that air cooling consumes more parasitic power than liquid immersion cooling, especially for high battery loads. Moreover, Moghaddam [131] simulated the cooling performance of a module with 21700 cells and concluded direct cooling with air and dielectric coolant results in the lowest temperature gradient inside the battery system. The disadvantage of air cooling is the parasitic power required and low heat capacity and challenging flow control. Immersion cooling with dielectric fluid was, therefore, concluded to have to greatest cooling performance. However, disadvantages include: the added complexity/cost of condensing evaporated vapour, potentially higher pumping losses in high viscosity fluids, high cost of the fluid, material compatibility issues and additional weight of the fluid.

Yet, despite these challenges, Immersion cooling has great potential

as a direct cooling method of BTMSs, however a consensus on which type of system and what fluid to use has yet to be determined. The following sections therefore explores single phase and 2-phase systems as well as efforts towards investigating the merits of using different working fluids.

4.1. Candidate dielectric fluids

Single phase immersion cooling fluids can come under several categories which include: hydrofluoroethers, hydrocarbons, silicon oils and water/glycol. Single phase immersion cooling has benefits over 2 phase immersion cooling, in that they tend to be less expensive both due to the liquid itself and the system used to contain them. The ease of implementation was highlighted by Varma [132] who compared Novec 7000, a 2-phase fluorocarbon-based fluid, to GRC Electrosafe, a single phase hydrocarbon based fluid, for a server system. They highlighted that both the single phase and 2 phase systems were more expensive than air cooling but achieved far superior heat transfer abilities which warranted the additional cost.

In immersion cooling systems, the fluid properties play a vital role in determining the cooling efficiency and thermal stability. There are several crucial requirements for immersion cooling fluids. Firstly, it must be electrically insulating due to the contact between the battery and fluid, which could provide a potential pathway for electron transport, and a low electronic conductivity will limit potential electronic leakage issues. Water/ethylene glycol-based coolants are popular coolants for indirect cooling systems. However, due to the conductive characteristics of water, the coolant is often not suitable for immersion cooling system except in certain unique designs where the batteries can be electrically isolated. Secondly, the fluid needs high specific heat capacity and thermal conductivity. Thirdly, fluids which are non-flammable or have sufficiently high flash points, are needed, to ensure safe operation and to reduce fire risk in case of a battery thermal runaway. Finally, the ideal fluid needs to be made available in large quantities, especially for passenger vehicles with mass production. In addition to the abovementioned factors, the proper working temperature range, longevity, good material compatibility, low weight, low viscosity, and sustainability are other critical aspects when it comes to select a suitable immersion cooling fluid type. In this section, typical immersion cooling dielectric fluids will be introduced with key metrics

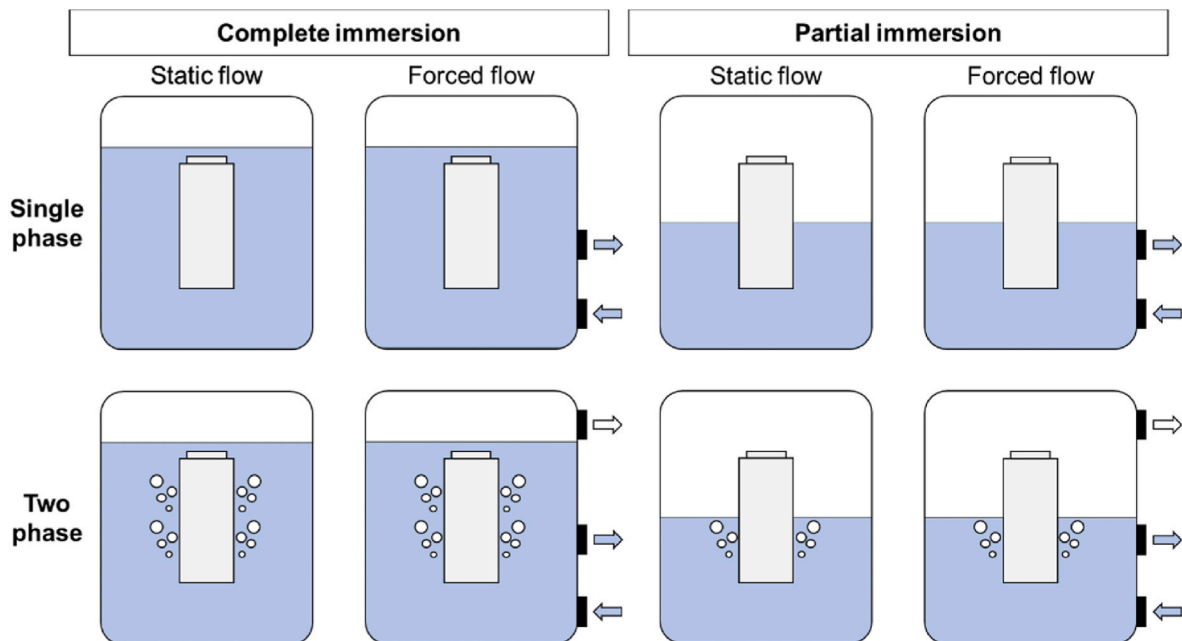


Fig. 6. Overview of the different types of immersion cooling considered for battery thermal management systems.

listed in Table 3.

4.1.1. Hydrofluoroethers

Hydrofluoroethers have received immense interest in the area of power electronics immersion cooling [142,143], but recently its working scope has been extended into BTMSs [144–146]. Novec engineered fluids are one set of hydrofluoroether products, developed by 3M, with a wide range of thermal properties. When the battery temperature increases to the boiling point, the fluids start to boil which enhances the heat transfer characteristics of the fluid considerably. Furthermore, 3M Novec products are non-flammable, which minimises some of the system safety concerns [73].

Recent works towards investigating hydrofluoroethers as working fluids in direct cooling systems includes van Gils et al. [147] who explored the applicability of Novec 7000 as a cooling fluid for a 18500 cylindrical battery. This has a boiling point of 34 °C making it ideal for use in BTMSs, though it was noted that this boiling temperature can further be controlled by adjusting the system operating pressure. In their work, during operation, the battery temperature did not rise more than 5 °C and was maintained below the fluid boiling point, while the equivalent air cooling system exceeded 40 °C. The homogenisation of cell temperature around the boiling temperature is therefore, a key advantage of these 2-phase systems, which leverages the large latent heat of vaporisation. The condensation of the resulting vapour, however, does often require an additional cooling system, but this could be integrated into the broader thermal system of an EV.

The effectiveness of this boiling process is, however, strongly temperature dependent and non-linear. Fig. 7 shows how the heat flux varies with temperature. The non-linear trend seen in the heat transfer properties was attributed to the varying physical mechanisms occurring in the system which are annotated in the figure. Up to point A, the fluid remains below its boiling point, and heat transfer is dominated by the liquid phase natural convection. Heat flux gradually increases with increasing temperature due to proportionally larger convective flows. From A to B, vapour phase nucleation begins on the surface of the cell, enhancing the surface heat transfer through the turbulent mixing caused by the bubble formation and movement. By point C, boiling nucleation has fully developed and peak heat flux has been achieved. After this point the bubbles forming at the nucleation points begin to agglomerate and starts to inhibit new boiling sites, leading to a decrease in the heat flux. After point D, a film of the vapour on the surface of the cell has formed leading to film boiling, where a layer of vapour insulates the cell. The trend in heat flux with temperature again continues to rise with temperature but with less efficiency due to the insulating nature of the gaseous film.

Another experiment related to benchmarking air cooling systems and immersion cooling systems was performed by Hirano et al. [148]. Here a battery module with 10 cells connected in series was submerged in Novec 7000, with the module cycled aggressively at 10C and 20C. For the air cooling system, the battery temperature reached 80 °C at 10C within 5 cycles and 90 °C at 20C after 2 cycles. Conversely, the immersion cooling system exhibited excellent thermal performance,

maintaining battery temperature at 35 °C with less than 1 °C difference under 10C cycling. This temperature stability was then maintained at 35 °C even at higher current of 20C, highlighting both the impressive thermal characteristics of hydrofluoroethers but also the non-linear heat transfer effects occurring in the system.

In the case of An et al. [144], they investigated using Novec 7000 on LFP power prismatic cells. Rather than immersing their battery module in Novec 7000, they used a microchannel cooling system which used plates to make thermal contact with the cell. In this arrangement the flow rate of the coolant could be finely controlled, which the authors also used to control the boiling point of the fluid. Wang et al. [146], on the other hand, had a slightly more elaborate Novec 7000 cooling system which they also modelled alongside the experimental results. They found that bubble nucleation was more effective for creating a uniform temperature distribution within the module due to the resultant turbulent flow. However, forced convection of the Novec 7000 in a single phase gave rise to a reduced maximum temperature of the battery module to 31.5 °C with a 5C discharge and an inlet velocity of 0.3 ms⁻¹. Whilst this is useful, this paper only investigates the uniformity of the temperature distribution at a pack level not cell level.

Clearly, complex heat transfer behaviour occurs during this transient boiling regime. Birbarah et al. [71] studied how the heat transfer coefficient varies between natural convection and nucleate boiling for a Novec 72DE system, which has a boiling point of 43 °C, similar to Novec 7000. During operation when natural convection dominated, the convective heat transfer coefficient was found to range from 2.0 ± 0.3 to 5.4 ± 0.5 kW/(m²K). During nucleate boiling this increased to between a range of 5.4 ± 0.54 to 20.8 ± 4.2 kW/(m²K). The effect of nucleate boiling thus increased the heat transfer coefficient by a factor of 10 compared to single phase cooling. The significance of this increase is highlighted in the comparison with air cooling which has a heat transfer coefficient of 10–100 W/(m²K).

The performance of these Novec based cooling systems, from the academic literature, is therefore promising, however no studies have investigated the lifetime performance of these systems, which represents a major gap in the literature. Also, despite these achievements, there are some barriers hydrofluoroethers to realize the EV mass production scale such as the material cost and disposal. In addition to that, hydrofluoroethers have a density approximately 40% greater than water-glycol systems, resulting in additional weight of the total system. Novec 7000 also has the potential to increase the safety of EVs due to its high flash point but more research is needed to understand what would occur under thermal runaway.

4.1.2. Hydrocarbons

Recently hydrocarbon-based fluids have received increasing attention for EV battery immersion cooling systems. In general, hydrocarbon fluids can include: mineral oils, poly-alpha-olefins (PAO) or other synthetic hydrocarbon oils. “Mineral oil” is a term used for a wide range of distillates, primarily from petroleum, which is one suitable candidate for battery immersion cooling systems due to the low cost, low toxicity, and adequate working temperature range [149]. For instance, Patil et al.

Table 3
Thermal and physical properties of various single phase fluids considered in BTMSs.

Material	Kinematic Viscosity at 20 °C (cSt)	Density at 20 °C (g/ml)	Thermal conductivity (W/mK)	Dielectric constant	Specific heat capacity (J/kgK)	Boiling point (°C)	Flash point (°C)
Silicone oil	994.2 [133]	0.97 [133]	0.15 [69]	2.75 [134]	1370 [69]	140 [133]	316 [133]
Water-Glycol (1:1) mix	4.9 [135]	1.08 [135]	0.40 [135]	64.92 [67]	3,473 [136]	107 [136]	111 [137]
Mineral oil	56.0 [63]	0.92 [63]	0.13 [63]	2.10 [72]	1,900 [63]		115 [138]
Shell thermal fluids	19.4 [139]	0.81 [139]	0.14 [139]		2,100 [139]		190 [139]
Poly-alpha-olefins (Chevron Phillips)	5.1 (@ 40 °C) [140]	0.80 [140]	0.14 [141]		2,241 [141]		159 [140]
Hydrofluoroethers (3M Novec 7000)	0.3 [73]	1.40 [73]	0.08 [73]	7.40 [134]	1,300 [73]	34 [73]	None [73]
Ester (MIVOLT DF7)	16.4 [68]	0.92 [68]	0.13 [68]		1907 [68]		194 [68]

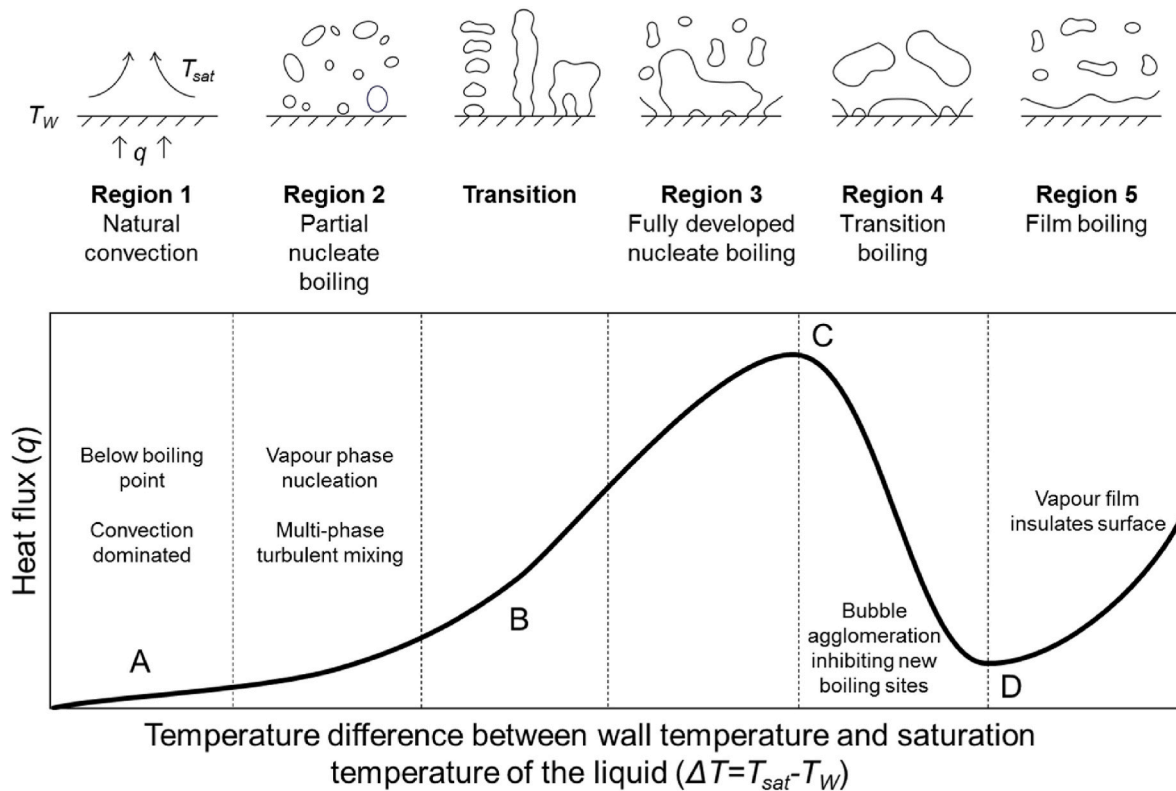


Fig. 7. Diagram showing the variation of heat flux (q) and nucleation as the difference between the wall temperature and the saturation temperature increases. Adapted from Van Gils et al. [147].

[150] investigated the performance of mineral oil as a cooling media for a pouch cell pack. For a single cell, the maximum battery temperature was maintained at 32.8 °C, 30.8 °C, and 30.6 °C for different flow rates of 1 L/min, 5 L/min, and 10 L/min, respectively. This showed whilst flow rate is a performance factor, increasing the flow rate from 1 L/min to 10 L/min only reduced cell temperature from 0.89 °C to 0.42 °C; suggesting lower flow rates give sufficient performance. Other considerations include the fact that as mineral oil is refined from crude oil, depending on the refining process, it still contains impurities which can subsequently lead to poor oxidation stability. Potential impurities include sulphur containing compounds which can be the cause of copper corrosion in electrical systems.

PAOs are another potential base material for EV battery immersion cooling fluids. Compared with mineral oil, PAO has similar features such as low toxicity, wide working temperature range. They contain more saturated chemical bonds between carbon atoms than mineral oils, which in turn delivers more a stable structure. Furthermore, the viscosity of PAO can be controlled over a wide range. For example, Chevron Phillips synthesized five various C12-based PAO products with kinematic viscosities at 100 °C ranging from 2 cSt to 9 cSt [151]. However, as of today, there are limited examples related to battery immersion cooling with use of PAO based fluids.

Shell have recently launched “Shell Thermal Fluids E5 TM” series which utilize Shell’s proprietary gas-to-liquid (GTL) base fluid technology, this opens another family of potential materials which can be used in immersion cooling systems. The GTL process produces base fluids which consist of mainly isoparaffines. These fluids show excellent thermodynamic properties and low weight compared to other fluid types and are virtually sulphur-free, which makes them non-corrosive against metals used in electrical systems. Hofer Powertrain modelled and tested these GTL fluids alongside other fluids in immersion cooled battery pack designs in a comparative study for ultrafast charging with conventional and modified indirect systems. Kreisel Electric recently reported abuse

tests performed with Shell thermal fluids and demonstrated that battery modules can be designed to mitigate the normal safety risks [152]. These fluids can also be used in other electrical BEV parts such as the electric motor and inverters; simplifying the powertrain.

Shell Thermal Fluids are compatible with many common materials, such as rubbers, plastics and metals which are representative materials applied in EV thermal management system. The shortcomings of Shell Thermal Fluids, however, include: could be flammable and have flash points (for instance, the flash point of Shell Thermal Fluid E5 TM 410 is 190 °C). It’s worth noting that increasing the fluid viscosity could increase the flash point. However high viscosities are not favourable in immersion cooling as low viscosities encourage turbulent flow and convection in an immersion system. For the time being there is not an agreement on the requirement and limit for flash point from EV manufacturers, which remains a gap academically and industrially.

4.1.3. Esters

Esters are another kind of dielectric coolant, which have achieved wide utilization due to its rapid biodegradability, low cost, high flash point, strong moisture tolerance, and favourable dielectric properties [153,154]. Esters can be divided into synthetic and natural. Natural esters are generated from vegetable oils with a glycerol backbone, while synthetic esters are generated from the reaction between polyol and carboxylic acids [155]. Compared with natural esters, synthetic esters offer superior oxidation stability that could reduce maintenance cycles. However, synthetic esters usually suffer from a relatively lower flash point compared to natural esters and often exhibit an undesirable odour. Reviews such as Mehta et al. [156] provide a useful comparison of natural esters and mineral oils as immersion cooling fluids in transformers, highlighting comparable breakdown voltages of >35 kV (2 mm gap) and viscosities of <30 and < 12 cSt (40 °C), respectively. The growing popularity of esters in thermal management of high voltage electronics was further highlighted by Ortiz et al. [154] who noted over

1.5 million devices being cooled by natural esters. However, they also showed that the cooling capacity of these organic fluids decreases with aging at elevated temperatures due to increasing viscosity, highlighting the need to understand the fluid behaviour over the lifetime of use. Thus, whilst esters have been demonstrated in the thermal management of high voltage electronics, with similar properties to mineral oils, their application in BTMSs remains a major gap. So far, only M&I Materials have launched ester-based immersion cooling dielectric fluids named MIVOLT, which includes a low viscosity product DF7 and a high viscosity product DFK [68] which are specifically for EV battery immersion cooling.

4.1.4. Silicone oils

Silicone oil is another candidate for immersion cooling. The viscosity of the silicone oil is dependent on the length of the chain of siloxane monomer, similar to hydrocarbons and has good temperature resistance at high and low temperatures because of its boiling point at 140 °C and melting point of −55 °C for a 5 cSt silicone oil [157].

Due to its dielectric capabilities, Silicon oil is a good candidate for immersion cooling in other energy sectors such as solar panels. Sun et al. [158] investigated and found that its optical transparency and low electrical conductance made it ideal for this application and can be directly transferred to LIB immersion cooling. Matsuoka et al. [159] compared different single phase immersion coolants: Novec 3283, Novec 43, 50 cSt silicone oil, 20 cSt silicone oil and soybean oil. They concluded that convective heat transfer dominates in many applications; therefore resulting in lower viscosity fluids providing superior performance. When comparing the 20 cSt silicone oil and the 50 cSt silicone oil, natural convection was more prominent in 20 cSt silicone oil.

4.1.5. Encapsulated systems with a water or water/glycol immersion fluid

Water/glycol is a mixture of different amounts of water and ethylene glycol. Many of the dielectric fluids considered for immersion cooling such as silicone oil or hydrocarbons have low thermal conductivities in comparison to water glycol. However, whilst the cost of water/glycol is favourable, the relatively poor electrical insulating properties remain a challenge for practical implementation. Yet, despite this challenge, authors have explored ways around this issue in other fields. Birbarah et al. [160], for instance, experimentally and theoretically compared water-glycol against other dielectric fluids in the thermal management of electronics. In order to insulate the circuit boards they coated it in Parylene C [161] and demonstrate how a layer as thin as 1 µm is able to provide sufficient electrical protection. Here, they found that the heat flux for a water-glycol system was up to 2x larger than that of the dielectric fluids tested. This was due to the increased thermal conductivity. The dielectric fluids they compared against were Novec 72DE and Novec 7300 where the boiling points of these dielectric fluids are 43 °C and 98 °C, respectively [74,162], whereas the boiling point of the 50/50 Water-Glycol was 107 °C [71]. For thermal conductivity, Novec 72DE, Novec 7300 and water-glycol had a stated values of 0.06, 0.063 and 0.4 W m^{−1}K^{−1}, respectively [163,164]. It should, however, be noted that although the water-glycol does have a greater thermal conductivity, the authors highlight that for lower power devices, the Novec 72DE may be a better option as its boiling point is lower and hence more heat would be absorbed through latent heat of vaporisation. This coating, and similar ones, may be applied to batteries to allow for fluids which are more electrically conductive to be used for immersion cooling. The use of a dielectric coating such as Parylene C for immersion cooling is therefore a promising approach, as it has been tested in air for 10 years at 80 °C without significant electrical, mechanical or thermophysical degradation in electronics applications [161].

A paper by Li et al. [165] investigated the use of a silicone and boron nitride sealant (SS + BN) to coat 18650 cells. 1C, 2C and 3C charging tests with both forced cooling and natural convection cooling was performed. Here, they found that the coating improves the thermal characteristics of the cell. The SS + BN was immersed in water was also able

to remove most of the heat generated by the cells in comparison to the other methods due to its higher thermal conductivity. They also state that this is also able to suppress non-uniform temperatures within the battery module as it was able to control the temperature to within 0.5 °C. Whilst maintaining a low temperature for a battery module is important to prevent thermal runaway, further investigation into temperature gradients at the cell level would be important to reduce degradation.

4.2. Sustainability issues relating to immersion cooling

The sustainability of the various BTMSs is also a critical consideration with works such as Lander et al. [166], providing a techno-economic comparison of the various approaches. Here they showed that battery life cycle costs could be reduced by 27% (from 0.22 \$/km for air cooling to 0.16 \$/km for surface/immersion cooling) with a 25% reduction in carbon footprint (from 0.141 kgCO_{2eq}/km to 0.104 kgCO_{2eq}/km, respectively). However, measuring the sustainability of an approach can be complex with the need to consider a wide range of factors. Blowers et al. [167], for instance, investigated the global warming potential of a range of hydrofluoroethers, with this being a combination of the substance's degradation rate in the troposphere and the amount of infrared radiation it absorbs. Here they highlighted the correlation between the higher number of C–H bonds and the lower atmospheric lifetime, which aligns with 3M's claims that their high molecular weight Novec fluids have limited ozone depletion potential. However, it is also noted that further data and research in this topic is needed for more conclusive findings, especially for emerging immersion fluids. Life cycle assessment of PCMs in building applications is however a much more researched area, with reviews such as Kylili and Fokaides [168] providing a potential framework for future studies. Here main categories of sustainability included: ecosystem, human health and resources.

Immersion solutions which use non-dielectric solutions such as that of Li et al. [165] who insulated their cells in a composite of silicone sealant and boron nitride with subsequent immersion into water, has the advantage of using environmentally friendly liquids. Here the sealant is essential to avoid electrolysis of the water however, approaches such as this might pose challenges in later recycling where brine solutions are sometimes used to safely discharge battery packs for recycling [169].

5. Battery safety

5.1. Immersion cooling safety

The purpose of the immersion cooling systems presently has not focused on mitigating the effects of TR [170], but on avoiding the conditions to TR. Battery TR typically is a result of excessive heat generation in combination with poor heat dissipation. Potential merits of immersion cooling include the potential to absorb large amounts of heat, especially in 2-phase systems, before more catastrophic mechanisms occur in the battery. Hydrofluoroethers therefore have favourable properties due to the latent heat of evaporation. However, if all the fluid was evaporated, or if insulating layers of gas form, this can affect the effectiveness of the system. Consequently, to enhance the heat transfer ability, the battery immersion cooling system should avoid the transition from full nucleate boiling to film boiling, where the heat removal capability is considerably hindered. This can be achieved with regulation of coolant flow rate or the operating pressure.

Examples demonstrating the increased safety characteristics of immersion cooled battery packs includes Zhou et al. [171] who immersed a NMC 622 pouch cell pack (3 cells with 60 Ah each) in Novec 649 which has a specific vaporisation heat of 88 kJ/kg and boiling point of 49 °C. To investigate the safety characteristics, they overcharged the middle cell of the pack at 1C. Here they noted that the use of the immersion fluid prevented the thermal propagation of the failed cell to adjacent batteries, limiting the impact of a single failed cell. Other notable efforts to

mitigate thermal runaway using phase change systems include that of Zhang et al. [172] who studied the TR behaviour of a hybrid PCM-liquid cooling system. They noted that in a pure PCM system, TR propagation between their 25 Ah cells was only delayed, and in an indirect aluminium cooling plate system, the high thermal conductivity of the cooling plates actually aided the thermal propagation. However, in a combined PCM-liquid cooling system, they noted that the combination of the thermal inertia of the PCM and the heat removal of the indirect liquid cooling was sufficient to prevent the propagation of TR. This highlights the need to consider the balance of high thermal conductivity during normal operation and its propagation during TR.

Alternative systems which have explored ways of mitigating the effects of TR include Larsson et al. [173], who investigated water mist emergency cooling for batteries ranging from 92 to 138 Wh with only 851 g of water, to delay the TR. The combination of liquid nitrogen, liquid argon and liquid carbon dioxide as immersion cooling agents was employed by Wang et al. [174], who defined the amount of latent heat of vaporisation to be 0.1–10 times of the TR heat generation. Bandhauer et al. [175] adopted the R125 refrigerant as a coolant and noted that 1 kg of liquid phase R125 can achieve up to 2 MW of cooling power, which can suppress TR within 0.1 s.

For fluids such as mineral oil, silicone oil and esters there have been similar fire safety tests done for use as a transformer fluid. Hellebuyck et al. [176] discuss some of the fire safety difficulties with immersing transformers in a dielectric immersion fluid. For the fluids investigated, the heat released per unit area was lowest for silicone oil. They also highlighted the difficulty in using small scale experiments to describe a full-scale fire and hence both small- and large-scale experiments are needed to fully assess the behaviour of the fire.

Detailed studies on the thermal and fire propagation of multiple batteries have revealed the triggering conditions, such as onset temperature and oxidizers. Battery immersion cooling can provide significant preventative measures to mitigate these threats [177–180]. Gao et al. [181], for instance, researched a design of an emergency refrigerant spray cooling thermal management system for a battery pack. The study indicated that refrigerant spray cooling has an obvious cooling effect and oxygen suppression performance. Liu et al. [182] carried out a numerical analysis of LIB TR with water mist suppression. The model simulated the cooling and suffocating effects of the cone angle, flow intensity, initial velocity and droplet diameter. From this, it was proposed that, when the battery ruptures, the flammable gas will vent, mixing with oxygen leading to flame ignition. One advantage for immersion cooling in preventing hazardous accidents, is that inorganic and organic coolant such as silicon oil, hydrofluoroethers and so on can dissolve this flammable gases and create barriers between fuels and combustibles [183,184].

5.2. Perspectives on battery safety enhancements

Personal safety remains paramount with the increasing deployment of EVs, with a specific focus on avoiding TR in EV accidents [180]. As temperature increases in a TR event, the battery's internal rate of chemical reaction will accelerate. The pressure of gas released by side reactions could, therefore, break the outer pouch or shell of the LIB. Once a gas-liquid-solid flammable mixture is ejected and reacted with oxygen, combustion will occur, potentially causing secondary disasters [185]. Thus, approaches which mitigate this are highly desirable with immersion cooling offering many advantages.

The lack of accurate understanding of the thermal hazards associated with reactive chemicals is a major cause for many fire and explosion accidents. As explained by Sun et al. [186], a TR process is generally governed by three factors: the heat generation due to abnormal reaction, the heat exchange with environmental, and the internal heat and mass transfer. To control thermal failure, the cooling method and emergency response are essential. Therefore, early detection of failure, including TR or fire, is crucial [187]. An accurate diagnostic analysis system coupled

with sensors of mechanical-thermal-electrical-chemical could warn when abnormal thresholds have been exceeded [180]. Also, compartmental design of battery systems should be considered where possible [188].

Taking into account the above factors into consideration, Zhang et al. [189] employed protection methods to prevent the occurrence, and mitigate the negative consequence, of TR within battery systems at both cell and system levels. At the cell level, safety vents, positive temperature coefficients (PTCs) and thermal fuses are used to prevent overpressure, which should be optimized. At the system level, more emphasis should be focused on the BTMS [190]. Wang et al. [180] summarized a three layer design concept of a BTMS with enhanced safety. Firstly, the system should ensure battery operation at the optimal temperature range. Secondly, it should detect critical points of mechanical-thermal-electrical-chemical failures and deliver alarm messages. Finally, once a thermal hazard occurs, emergency measures should suppress TR propagation effectively. Furthermore, Bravo Diaz et al. [188] highlighted the need to consider battery TR compartmental design, detection and suppression methods also. Compartmental design is to hinder thermal propagation, and can be achieved by increasing the cell spacing [191] or adding thermal barrier materials between cells [192]. Detection methods can be achieved with a mix of terminal voltage, temperature, gas and structure deformation detection of the battery. The suppression approaches should include smothering, cooling, chemical suppression, or isolating the fire [193,194], of which immersion cooling has demonstrated key advantages in.

Therefore, reflecting on this, we recommend that thermal safety designs should take five factors into consideration, including standard manufacturing and mounting, understanding thermal properties, diagnostic analysis sub-systems, thermal control sub-systems and emergency response sub-systems, which are articulated in Fig. 8.

6. Industrial insight

In this section, we examine the existing applications of battery immersion cooling to EVs and energy storage. As this section speaks to the industrial application of immersion cooling, most of the content focuses on conference presentations and webinars, which reflects the fact that many organization hold IP in this space and do not publish their findings in the scientific literature. As we show, there have been a number of convincing proof-of-concepts. To date, we have already identified more than 10 EV system suppliers that have achieved significant progress with battery immersion cooling.

Recently, AVL presented information on how they overcame technical limits (timing, cost, and complexity) when implementing battery immersion cooling [195]. The weight and cost of battery module was largely reduced by cooling electrical connections directly. Meanwhile, the safety was also improved, with benchmarks made against the standard ECE-R100, UN Transportation and GB-T 31467-3 tests.

Ricardo Engineering [196] showcased a “turn-key” battery module with immersion cooling technology. Using a 21700 cylindrical cell-based module with M&I MIVOLT fluid, a high charging rate of 3.9C was achieved. It was found that this approach meant that the maximum battery temperature could be controlled to around 30 °C. In contrast, it was shown that the same batteries would reach temperatures of up to 50 °C when the same C-rates were applied within a BTMS using cold plate cooling. Furthermore, they explored the module's safety through thermal runaway tests. It was found that despite triggering a cell into TR, the temperature of adjacent cell was maintained at 70 °C, which offers an attractive safety improvement over air cooled modules. They also mentioned that in contrast to cold plate indirect cooling, the cost of immersion cooling can be reduced by 8% at module level and 6.5% at vehicle level. This was attributed to several factors with the main one being the cost savings from the thickness reduction of copper busbars.

Kreisel Electric have developed an efficient and safe battery solution combining Kreisel's laser-welded battery module that includes single

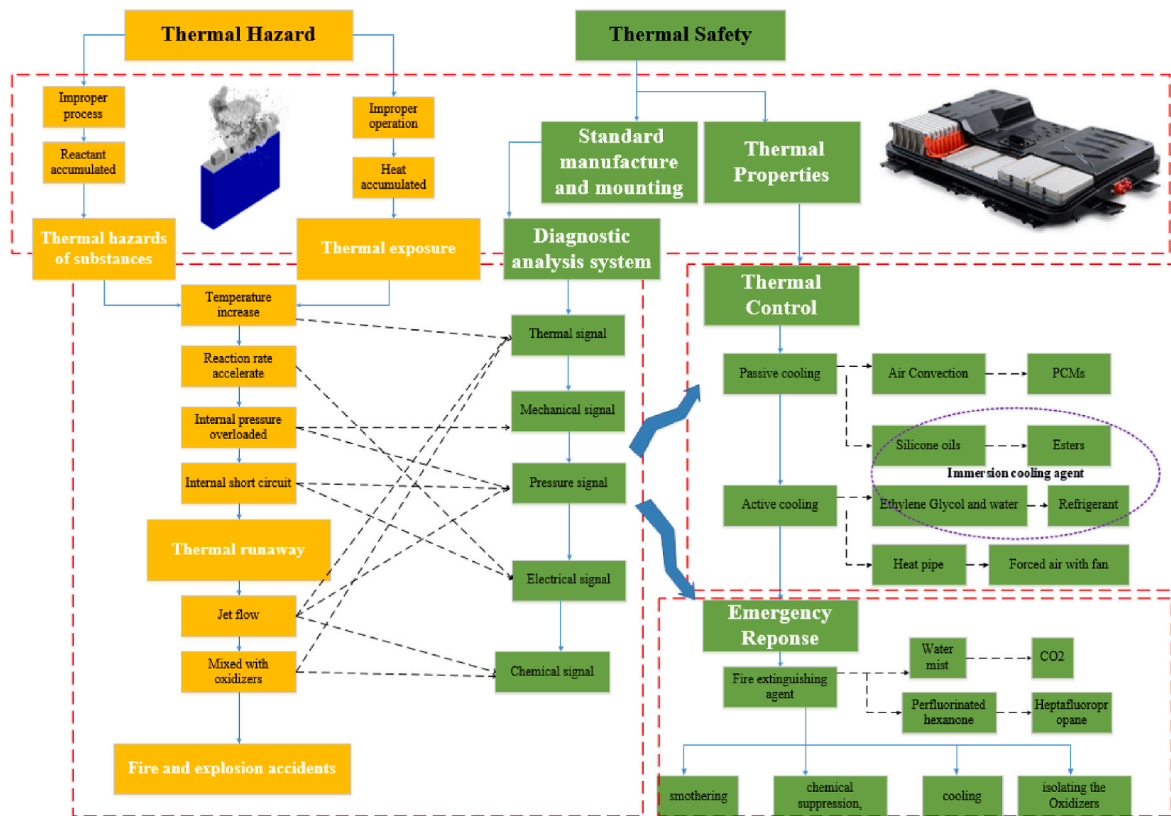


Fig. 8. Proposals for battery safety enhancement.

18650 or 21700 cylindrical cells, fusing, and controlled de-gassing channels with Shell's proprietary GTL based thermal management fluid, which immerses the battery cells [197]. The Kreisel battery system is already installed in commercial vehicles, such as the London electric bus fleet, industrial applications such as concrete mixer built by Liebherr, and high performance charge posts such as the Chimero battery backed EV Fast Charger built by Kreisel electric. In addition, Kreisel battery immersion cooling will be used in motorsport applications e.g. as a battery platform for the Federation Internationale de l'Automobile (FIA) world rally championship hybrid series, and as a technology platform for the FIA World Rally Cross all electric 1e series from 2022.

Xing Mobility [198,199] has developed battery immersive cooling technology through their IMMERSIO™ battery module system. The Xing mobility solution has been proposed in mining, construction, and agriculture applications. Meanwhile, e-Mersiv and Exoes are technology companies based in France who have developed an immersive BTMS with charge speeds upto 4C for all cell formats (cylindric, pouch and prismatic) [200,201].

Some proprietary technologies have also been investigated though they have yet to be put into real-world application. Tesla patented a "battery coolant jacket" describing a battery module with an integrated frame structure to hold battery cells which are surrounded and cooled directly by a liquid [202]. Anhui Xinen Technology Co describe in a patented battery module and pack design with increased contact areas between coolant and battery surface, thereby improving cooling and safety of the battery [203]. LION Smart GmbH developed a light-weight battery pack with integrated immersive cooling technology using 3M Novec fluids, which can be used in automotive or aviation applications. LION Smart are participating with this technology in the EU funded LIBAT Clean Sky 2 Project aiming towards climate neutral aviation by 2050 [204].

Battery immersion cooling is finding applications in high end luxury sports cars. The new McLaren [205] "Ultimate Series" named as

"Speedtail" is the first serial car worldwide to implement immersive battery technology. The batteries within this car are permanently immersed in a lightweight dielectric fluid to improve thermal management, allowing the cells to deliver greater performance for longer. McLaren claimed that the Speedtail can generate the total output of 1055 horsepower while the battery pack provides a power density of 5.2 kW/kg [205].

In addition to the McLaren supercar, battery immersion cooling has also been launched in the EV retrofits of classic sports cars. In the product specification of their conversion of the classic Alfa Romeo GTV into a high performance BEV, Totem Automobili highlighted their use of 3M Novec to immersion cool the battery back, delivering greater performance for this vintage classic, ensuring its design legacy will last long into the future [206].

Meanwhile, Finland based Moveko developed a 120 V DC drivetrain system for public transportation fleet operators including an immersion cooled modular battery system suitable for high demands and reliability. The full systems enable high power ultrafast charging in combination with contactless charging technology [207]. Finally, AMG launched in 2021 a new plug-in hybrid technology introducing an immersive battery system adapted from the Daimler Formula 1 race car. The HPB80 is a 400 V battery with 6.1 kWh capacity, 70 kW constant power and a peak power of 150 kW which is available for a few seconds [208].

7. Conclusions

In this review, battery thermal management methods including: air cooling, indirect liquid cooling, tab cooling, phase change materials and immersion cooling, have been reviewed. Immersion cooling with dielectric fluids is one of the most promising methods due to direct fluid contact with all cell surfaces and high specific heat capacity, which can be increased even more if the latent heat of vaporisation is used in 2-phase operation. Such high heat rejection capability can also improve

safety by suppressing thermal runaway under some circumstances. A controllable boiling point has also been demonstrated as an effective means of homogenising cell temperatures in a pack. However, both single phase and 2-phase immersion cooling come with additional integration challenges. Furthermore, the heat removal ability of 2-phase immersion cooling varies non-linearly with temperature, due to the complex nature of the nucleate boiling transition to film boiling. Gaseous films forming around cells have been found to insulate cell surfaces and thus needs to be carefully considered during system design. Nonetheless, the heat removal capabilities of single-phase immersion cooling ($2\text{--}5\text{ kW m}^{-2}\text{K}^{-1}$) can be 2 orders of magnitude higher than air cooling air cooling ($10\text{--}100\text{ W m}^{-2}\text{K}^{-1}$) with 2-phase immersion cooling ($5\text{--}20\text{ kW m}^{-2}\text{K}^{-1}$) potentially reaching 3 orders of magnitude higher.

Cooling fluids suitable for immersion cooling include: hydrofluoroethers, hydrocarbons, esters, silicone oils and water-glycol mixtures. Key figures of merit include viscosity, density, thermal conductivity, dielectric constant, specific heat capacity, boiling point, flash point and cost. Of these, the hydrofluoroethers have shown significant promise, with the 3M Novec series of fluids finding application in BTMSs. Hydrocarbon based fluids have also shown promise, however relatively little research has been conducted on its performance as a BTMS working fluid. Whilst non-dielectric fluids such as water-glycol have the drawback of poor electrical insulating properties, authors have explored various coating methods on cells to mitigate this and thus they remain a consideration for future BTMSs.

Based on this literature review the following research gaps have been identified which would benefit from further investigation. This includes: the impact of immersion cooling on the lifetime of different LIBs, understanding low temperature performance, studies on the long-term material compatibility/stability of immersion cooled BTMSs, safety studies and sustainability of the candidate fluids.

Yet, despite these challenges, immersion cooling systems remains a promising approach for battery thermal management, with this review paper provides insights into the thermally coupled behaviour of batteries and the potential of different working fluids.

Declaration of competing interest

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