Review

Research Progress of Shear Thickening Electrolyte Based on Liquid–Solid Conversion Mechanism

Qianqian Huang [1], Xin Liang [1,*], Bing Liu [2,*] and Huaxia Deng [2]

[1] School of Energy Materials and Chemical Engineering, Hefei University, Hefei 230601, China; huangqianqian@stu.hfuu.edu.cn

[2] CAS Key Laboratory of Mechanical Behavior and Design of Materials, Department of Modern Mechanics, University of Science and Technology of China (USTC), Hefei 230027, China; hxdeng@ustc.edu.cn

[*] Correspondence: x.liang@hfuu.edu.cn (X.L.); bingliu@ustc.edu.cn (B.L.)

Abstract: As an essential component of the lithium-ion battery system, electrolyte plays a crucial role in ion transport between the electrodes. In the event of thermal runaway, commercial organic electrolytes are prone to internal disturbances and fires; hence, research on safe electrolytes has gradually become a hot topic during recent years. Shear thickening electrolyte, as a new type of smart electrolyte, can exhibit a liquid state in the absence of external force and rapidly converts to a quasi-solid state once the battery is subjected to drastic impact loading. In this paper, the recent progress of shear thickening electrolytes with liquid–solid switching performance is presented, including its working principles, synthesis and preparation procedure, and battery performance. Additionally, the perspective and challenges for practical application are discussed.

Keywords: shear thickening electrolyte; liquid-solid reversible conversion; safe electrolyte; STF

1. Introduction

Lithium-ion batteries (LIBs) are playing an important role in the fields of cell phones and computers, new energy vehicles, and aerospace and have attracted extensive research and application worldwide. For example, it was recently reported that globally the number of new plug-in electric vehicles will be nearly 14 million by 2023, about 35% higher than in 2022. However, with the largescale popularization and application of electric vehicles, the collision safety performance (mechanical abuse) gradually captured the attention of researchers around the world [1,2].

Commercial lithium-ion batteries are generally composed of a porous polymer separator, an electrolyte, and positive and negative electrodes [3]. In recent years, there have been numerous reports of explosion and fire accidents in new energy vehicles. These accidents are typically caused by internal thermal runaway caused by mechanical abuse. When a battery is subjected to an external shock, the interaction of oxygen, heat, and fuel is likely to trigger a fire or explosion, which poses a threat to human life safety. The thermal runaway process of the battery is shown in Figure 1 [4]. When the battery is subjected to external high-speed impact, the internal short circuit is triggered due to the rise in temperature and the thermal contraction of the separator, which eventually leads to thermal runaway of the battery [5]. To solve the problem of explosion and/or fire caused by mechanical abuse in battery systems, flame retardant additives, physical protection packages, and stable electrolytes (mainly including solid-state electrolytes and ionic liquid) have commonly been used to improve its safety performance [6]. Organic phosphate compounds are the most commonly used flame retardant additives, including phosphate ester, phosphite ester, and cyclophosphonitrile, among which phosphate ester flame retardants include trimethyl phosphate (TMP), triethyl phosphate (TEP), tributyl phosphate (TBP), and triphenyl phosphate (TPP) [7]. The addition of flame retardant additives is beneficial to the flame-resistant properties of electrolytes; however, this also leads to an increase in the viscosity of the electrolytes and a decrease in electrochemical

performance [8,9]. Xiang et al. [10] found that good electrochemical compatibility between DMMP flame retardant additive and graphite anode can be achieved by adding film-forming additive VEC or surface modification. However, with continuous cycling, electrochemical impedance becomes larger, eventually leading to continuous attenuation of battery capacity. Another method is to add physical protection packs, which increase the weight and additional burden on the battery [11,12]. The advantage of ionic liquids is attributed to their nonflammability. However, the disadvantages include low multiplicative performance, low lithium transfer numbers, and high costs, problems which need to be solved [13,14]. Traditional protection methods are usually at the cost of partial electrochemical performance and unnecessary waste. Therefore, exploring new electrolytes to build safer and more sustainable Li-ion battery systems has become a primary challenge for future development.
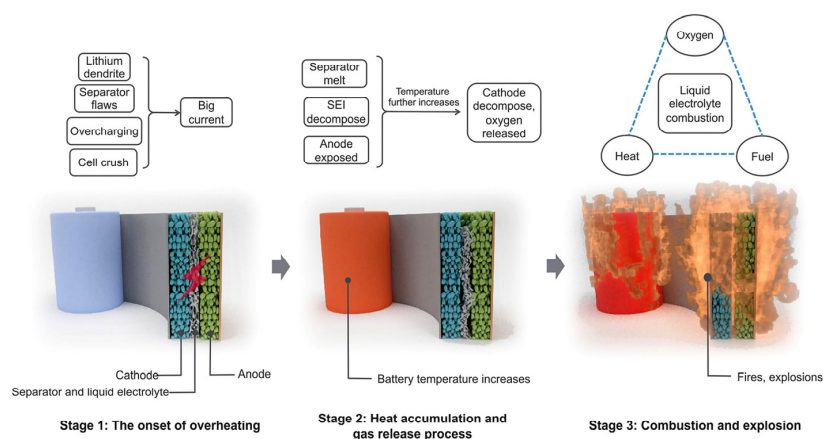

Figure 1. Three stages for the thermal runaway process. In the first stage, the battery gradually turns abnormal and the system temperature rises continuously; in the second stage, the battery temperature rises abnormally and an exothermic phenomenon emerges; finally, the flammable and evaporable electrolyte comes into contact with air and the system undergoes thermal runaway at high temperature and triggers a direct fire or even an explosion [4]. Copyright 2018, Science Advances.

Shear thickening electrolytes are prepared by dispersing inorganic uniform and solid fillers into the liquid electrolyte, which behaves like a liquid under normal operating conditions and performs nonlinear shear thickening performance upon external rapid shear rate [15]. The electrolyte exhibits quasi-solid behavior when the shear thickening performance is triggered, showing the high safety property of the battery [15]. Through the liquid–solid switching performance of this type of intelligent electrolyte, the battery avoids the risk of short circuit and thermal runaway under mechanical abuse conditions. In 2006, Zhang et al. [16] and Haregewoin et al. [17] reviewed the performance improvements of electrolyte additives for lithium-ion batteries over the past 20 years and provided an outlook on how to enhance battery safety. Kalhoff et al. [1] studied the additives used in electrolytes, and the results show that the additives are effective in preventing the overcharging of the thermal abuse of batteries. Shu et al. [14] recently described the development of shear thickening electrolytes (STE), and the results show that STE plays a key role in enduring mechanical abuse. Designing and developing this type of electrolyte can provide an effective approach to achieving high performance and safe lithium batteries.

2. Shear Thickening Mechanism

Shear thickening is a type of non-Newtonian behavior in which shear stress increases remarkably with increasing shear rate [18–20]. In Figure 2, the whole process of shear thickening is clearly shown. In the first stage, the system stays in the steady state; in the second stage, as the shear stress increases, the solid particles develop a layered structure and gradually lead to the shear thickening phenomenon. Finally, when the external shear rate exceeds the critical shear rate, the aggregation of solid particles leads to the shear thickening phenomenon; when the external shear stress is removed, the system shifts

back to the steady state [21–23]. The most common shear thickening system consists of silica particles and a solvent with a hydroxyl group (-OH) [24]. Raghavan et al. [25] found that the expanded silica particles were able to dissolve uniformly and undergo a shear thickening effect by adding the silica particles to a solvent with a hydroxyl group (-OH). The rheological behavior of the gaseous silica dispersed in different solvents was systematically investigated, and it was concluded that solvents containing methyl end groups (-methyl) lead to worse solubility and shear thinning behavior.
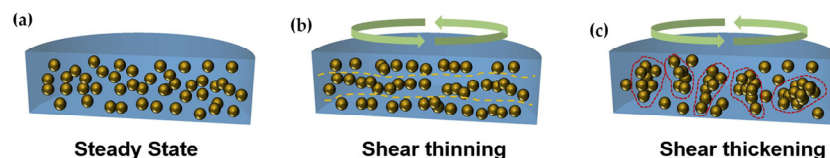
Figure 2. Schematic diagram of the process of shear thickening electrolyte in three states: steady state, shear thinning, and shear thickening. (**a**) First of all, under a steady state, the filler is uniformly dispersed in the fluid. (**b**) Secondly, owing to shear stress, the filler forms a layered structure, and the shear thinning behavior of the fluid is tentatively observed. (**c**) As the shear stress increases, the filler forms further water clusters, which eventually leads to a sharp increase in viscosity. When the stress is removed, the system shifts back to the steady state [23]. Copyright 2019, Elsevier.

Therefore, shear thickening fluid (STF) provides a unique material property to meet the needs of practical applications, including liquid body armor [26] and shock absorbers [27,28]. For example, Zhang et al. [29]. developed a type of C-STF-based yarn using carbon nanotube CNTs, hollow silicone rubber tubes, polydimethylsiloxane (PDMS) layers, and STF, which is a multifunctional smart yarn with excellent energy dissipation properties, responding to a wide range of mechanical stimuli such as bending, compression, and tensile, and possessing excellent impact resistance and impact detection. Liu et al. [30] used mesoporous silica (M-SiO$_2$) as the dispersed phase and ionic liquid as the solvent to prepare CNTs/STF/Kevlar smart materials with anti-impact, nonflammable, and sensing performance. Mesoporous silica is nanostructured with low thermal conductivity, and ionic liquids are green solvents with noncombustibility, impregnating fabrics in shear thickening solutions prepared from M-SiO$_2$. Ionic liquids can improve the high-speed impact resistance and flame-retardant properties of fabrics and significantly improve safety issues. Zheng et al. [31] fabricated MXene/cotton fabric (MCF)-based pressure sensors by means of which a variety of human health signals can be detected and differentiated, such as wrist pulses, early Parkinson's static tremors, and finger movements. Recent studies have pointed out that ballistic testing was performed to initially assess the effectiveness of shear thickening fluids in mitigating the effects of ballistic impact [24]. As shown in Figure 3a,b, the impact depths of the Class II and Class III armors with shear thickening behavior are shallow, at 6.1 mm and 18.55 mm, respectively, which are significantly lower than the other two types of armors. The same difference can be seen in the pressure film test results, indicating that the shear thickening fluid provides protection to the body. STF has unique physicochemical properties that provide significant possibilities for a broad range of applications in science and engineering [32], such as damper systems, medical equipment, and liquid body armor [26,33–35]. For example, Wu et al. [36] dissolved poly (vinyl alcohol) (PVA) in ethylene glycol (EG) and mixed it with STF to obtain a high-performance organogel electrolyte (PVA/STF), as shown in Figure 3c. This organogel electrolyte was applied to supercapacitors (SSCs), and the results showed that the highest ionic conductivity occurred at 80 °C. As the temperature continues to rise, the IR drops from 0.21 V to 0.075 V, which is a 2.8-fold decrease (Figure 3d). In Figure 3e, we can see that under external conditions such as bending, stretching, compression, and a high temperature of 80 °C, higher capacitance retention is shown only at a high temperature 80 °C; this indicates that ion mobility can improve and electrochemical performance can be enhanced under these conditions, resulting in better capacitance performance [37,38]. The energy dissipation capability of the SSC is significantly higher than that of the PSC due to the

energy dissipation characteristics of the STF during external impact loading, indicating that the device has good impact resistance to protect the body [39,40].
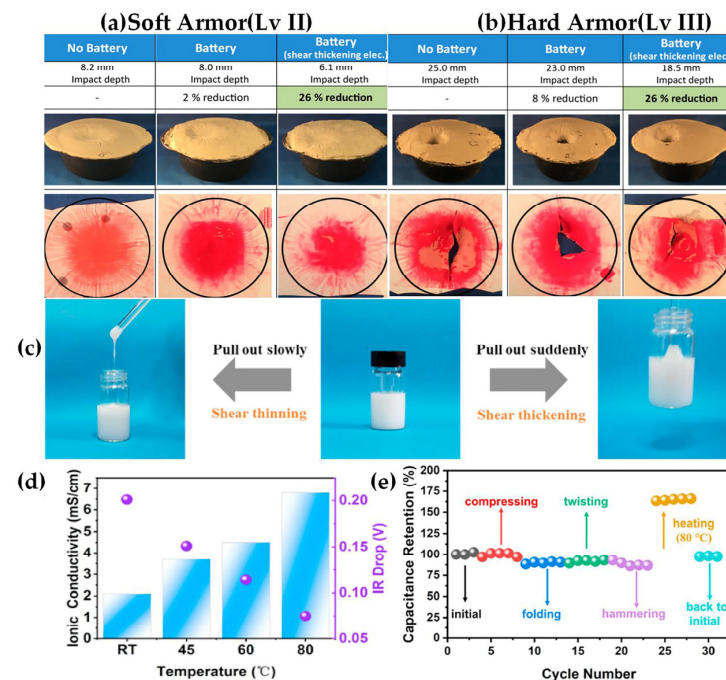
Figure 3. Applications of shear thickening effect: ballistic test results of (**a**) soft armor (Lv II) and (**b**) hard armor (Lv III) [24]. Copyright 2018, American Chemical Society. (**c**) Deepening the glass bar into the solution: when the glass bar is pulled out quickly, the solution demonstrates shear thickening behavior and turns to a solid state, which leads to the glass bar and solution sticking together; contrarily, when the glass bar is pulled out slowly from the solution, a shear thinning effect occurs. (**d**) Plots of ionic conductivity and IR drop at 25, 45, 60, and 80 °C. (**e**) Capacitance retentions of SSC under various external stimuli [36]. Copyright 2021, Elsevier.

STF performs conventional performance in static state, and particle agglomeration is quickly converted into a solid-like state under the high-speed impact, which plays an energy dissipation role. When the external force is withdrawn, the liquid performance is again carried out. This ingenious property will have infinite possibilities in the future if it is applied to smart devices.

3. Shear Thickening Electrolyte

Mechanical abuse refers to the occurrence of fire or explosion caused by external physical shock leading to short circuits and electrolyte leakage inside a battery. Research on safe lithium-ion battery electrolytes has focused primarily on solid electrolytes and ionic liquid electrolytes [41]. However, both inorganic solid electrolytes and ionic liquid electrolytes face significant challenges in practical applications [14]. For example, inorganic solid electrolytes are still deficient in terms of electrode interface stability and ionic conductivity [42]. Furthermore, ionic liquid electrolyte faces challenges such as low-rate performance, low lithium-ion migration number, and high cost [13,14]. As a result, to address the mechanical abuse issue, colloidal particles can be added to an inert dielectric solvent to form shear thickening electrolytes (STEs) [21]. The shear thickening effect is applied to the battery system, which can improve the comprehensive performance of the battery. Shear thickening electrolytes combine the strengths of solid and liquid electrolytes with excellent ionic conductivity and electrode compatibility; furthermore, they will be solidified under high shear, which protects against short-circuiting and combustion under physical shock. Upon mechanical pressure or impact, shear thickening electrolytes exhibit a shear thickening effect with an increase in viscosity, thereby dissipating the impact en-

ergy to protect separators and demonstrating tolerance to crushing. Thus, well-designed multifunctional electrolytes can significantly protect batteries at the early stage of battery thermal runaway [4].

It has been reported that ceramic particles play a critically vital role in various types of electrolytes [43,44]. In polymer electrolytes, adding ceramic particles such as SiO_2 [45,46], Al_2O_3 [47], TiO_2 [48,49], and AlSi [50] can enhance ionic conductivity and prevent crystallization. Here we illustrate this process with plasmonic conductors in a fuel cell. For example, solid polymer ceramic composites used in fuel cells exhibit favorable water retention properties, thermal stability, enhanced electrical conductivity, improved mechanical properties, and intentional molecular permeability [51,52]. Inspired by this, Ding et al. [53] prepared an electrolyte with shear thickening effect firstly by incorporating inert SiO_2 nanoparticles into an organic electrolyte in 2013. The experimental conclusions indicate that by comparing the viscosity and ionic conductivity of silica particles with different weight ratios (0, 6.3, 9.1, and 10.7 wt.% SiO_2 nanoparticles in 1M $LiPF_6$ + EC/DMC), it was found that the composite electrolyte with 10.7 wt.% silica had the best shear thickening effect, significantly higher than the other three groups of electrolytes. In addition, the ionic conductivity reached 1.93×10^{-3} S cm^{-1}, as shown in Figure 4a,b. When 9.1% fumed silica particles were dispersed into the electrolyte, the cell test results showed that the multiplicative performance was slightly higher than that of the conventional electrolyte. Additionally, there was only a slight change in electrochemical impedance before and after the impact test, which suggests that the shear thickening electrolyte did not negatively affect the electrochemical performance, as shown in Figure 4c,d. This study demonstrates that the electrolyte system under such a liquid–solid conversion mechanism has the capacity not only to preserve the basic electrochemical properties but also to improve the safety of the battery system.
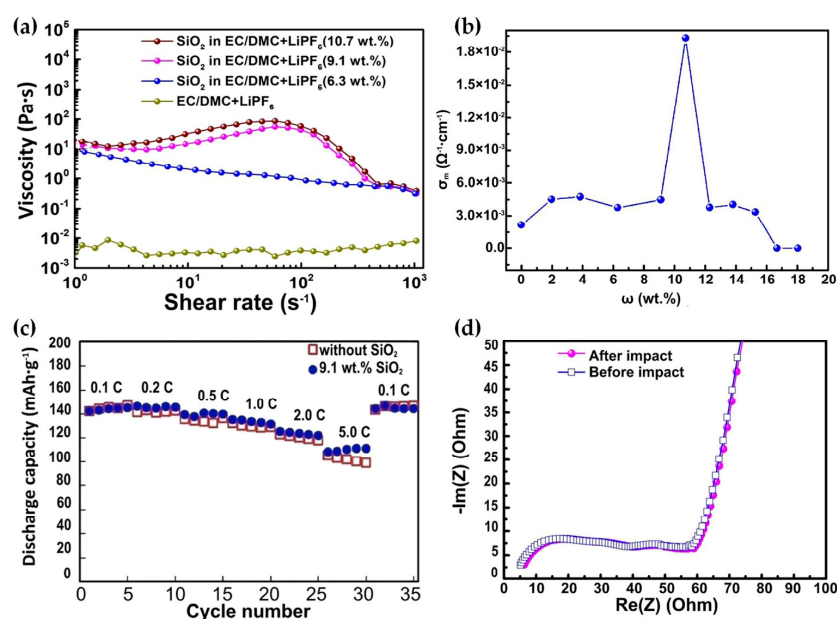
Figure 4. Electrochemical performance and viscosity test results: (**a**) Rheograms of electrolytes without SiO_2 and composite electrolytes containing 6.3% SiO_2 9.1%, SiO_2, and 10.7% SiO_2. (**b**) Plot of ionic conductivity versus mass fraction of fumed silica for composite electrolyte (SiO_2/$LiPF_6$/EC/DMC) at room temperature. (**c**) Under $LiFePO_4$ electrode: comparison of the rate performance of composite electrolyte (9.1% wt. fraction of SiO_2) with normal 1M $LiPF_6$ in EC/DMC electrolyte, (**d**) Electrochemical Impedance Spectroscopy (EIS) of graphite electrodes with STF before and after the impact test [53]. Copyright 2013, Nature.

While commercial organic electrolytes exhibit stable electrochemical performance, the safety of the system deserves attention due to the risk of flammability [54]. It has

been demonstrated that the addition of inorganic solid fillers to organic electrolytes can improve the safety of the electrolytes, but the electrochemical stability will be affected by the increasing proportion of fillers [24]. Therefore, it has become a hot research topic to appropriately reduce the proportion of filler that achieves shear thickening effect while maintaining the electrochemical stability of the system. Currently, in addition to spherical nanoparticles, it is noted that nanorods as well as fiber-based fillers have larger aspect ratios [55]. With an increasing aspect ratio, the critical volume fraction triggering the shear thickening performance decreases, and the challenge is to maximize the shear thickening effect at the lowest particle loading [56,57]. Ye et al. [24] dispersed silica nanorods with different aspect ratios into 1 mol/L LiTFSI + EC:EMC (1:1) and found that the volume fraction of filler that showed shear thickening behavior for silica nanorods with an aspect ratio of 24 was 0.146, while the critical fraction of silica nanorods with an aspect ratio of 5 was 0.358, which was higher than the former. As shown in Figure 5a,b, the rheological state diagrams indicate that nanorods with higher aspect ratios may provide discontinuous shear thickening at lower volume fractions. The results of the full cell assembled with nanorods with an aspect ratio of 5 show that the capacity of the cell continuously decreases as the cycle number increases at different multipliers, but the Coulombic efficiency remains stable. This may be attributed to the combination of water-etched poles on the silica surface, as shown in Figure 5c,d.
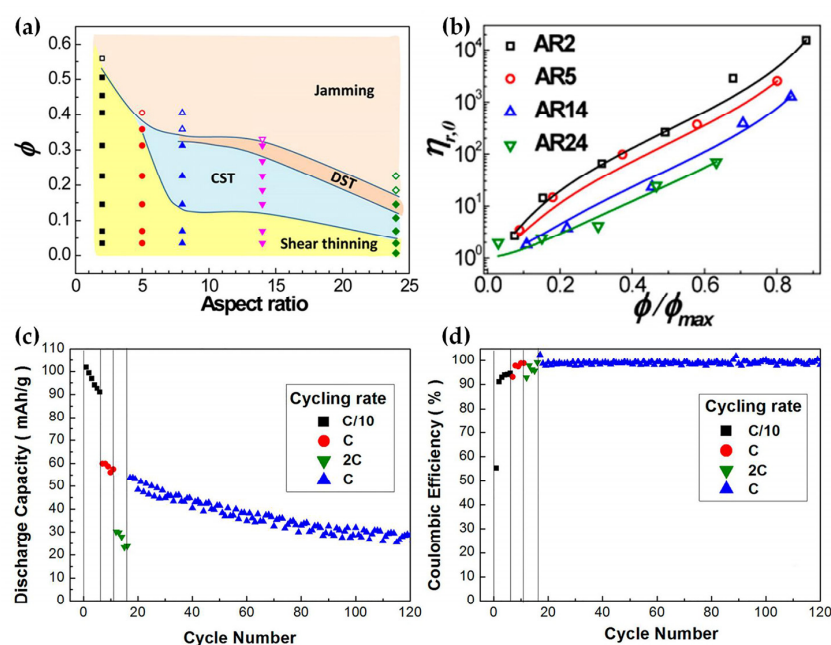
Figure 5. Performance comparison results of fillers with different aspect ratios: (**a**) rheogram of electrolytes containing silica nanorods with different aspect ratios and volume fractions. (**b**) Shear viscosity versus reduced volume fraction (Φ/Φ_{max}) of nanorod suspensions containing an aspect ratio of 2, 5, 14, and 24 at a shear rate of $1\,s^{-1}$. (**c**) Plot of the cyclic discharge capacity and (**d**) Coulombic efficiency of SiO_2 nanorods (33%content) with an aspect ratio of 5 in NMC|Graphite cells at different rates of multiplication [24]. Copyright 2018, American Chemical Society.

As described by Hamaker's theory, STF is a stable suspension in which the interaction forces between particles are universally stable [58,59]. Due to the weak interparticle forces ($\approx 10^{-10}$ N) of inorganic solid particles, surface functionalization of filler particles is necessary to support the formation of stable suspensions [23,60]. The complex composition of the electrolyte, which contains lithium salts, solvents, and additives, makes it more challenging to uniformly disperse the solid particles in the electrolyte. Additionally, the ionic conductivity is influenced by the volume fraction and needs to be explored concurrently. Based on the study of material modification and the regulation of an aspect ratio, Liu et al. [23] developed an electrochemically stable shear thickening electrolyte

using functionalized glass fibers (mGFs) interacting with the electrolyte to improve the safety of LIBs. Due to a larger aspect ratio, the critical volume fraction ratio of the filler to trigger the shear thickening behavior is 0.286. The glass fiber column was modified with (3-aminopropyl) triethoxysilane, and the material mGFs were synthesized as shown in Figure 6a. The amine end groups of the mGFs are able to interact with solvent molecules in the electrolyte, such as dimethyl carbonate (DMC) and ethylene carbonate (EC), forming a solventized layer around themselves. As shown in Figure 6b, the mGFs were added to the traditional electrolyte to obtain a uniformly dispersed shear thickened electrolyte, which is important for the system. The shear thickening effect was observed when the volume fraction reached 28.6%. High-speed impact tests have shown that the shear thickening electrolyte can withstand external impacts at 1.1–79 m s^{-1}, which proves that the electrolyte concentration increases instantaneously under greater shear forces. The electrochemical performance of the composite electrolyte was well maintained. The capacity retention rate of the LFP-Li half-cell after 500 cycles and the capacity retention rate of the LFP-LTO full-cell after 100 cycles were 95.2% and 94.4%, as shown in Figure 6c,d, respectively. It is worth noting that the application of shear thickening electrolytes is expected to replace commercial electrolytes by maintaining the electrochemical stability of the battery on the one hand and improving its shock resistance on the other hand to protect the battery from the hazards caused by severe shocks.
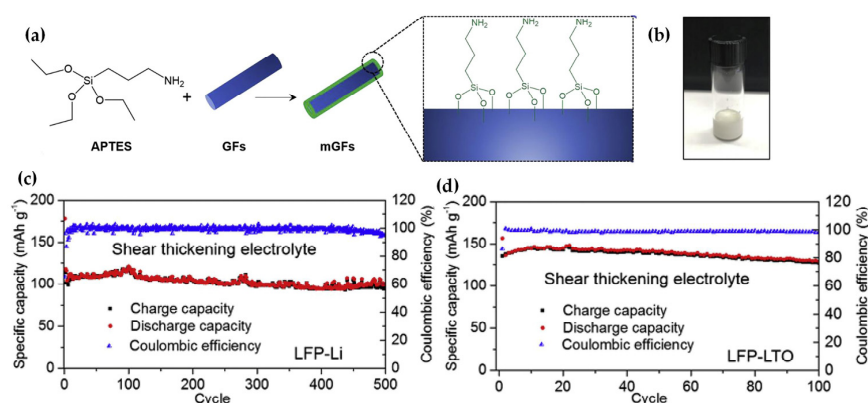
Figure 6. Synthesis and electrochemical properties of materials: (**a**) Synthesis process of mGFs (**b**) Uniformly dispersed shear thickening electrolyte in a clean vial. (**c**,**d**) 500 and 100 cycles performance of shear thickening electrolytes in LFP-Li and LFP-LTO cells [23]. Copyright 2019, Elsevier.

To further improve the application of shear thickening electrolytes, we gradually extended our study to other types of batteries. By preparing materials with different components, it was demonstrated that shear thickening electrolytes can improve the safety of batteries and avoid the safety hazards such as fire and explosions encountered in commercial electrolytes. For ZIBs, the formation of zinc dendrites are promoted due to uneven electrodeposition on the negative electrode, leading to a short circuit [58,61–63]. To alleviate zinc dendrite growth, Dong et al. [58] dispersed 55 wt% cornstarch in a 3 M ZnSO$_4$ and 0.2 M MnSO$_4$ aqueous electrolyte to obtain STE. As shown in Figure 7a,b, cornstarch particles are relatively monodisperse, although irregular in shape. When shear stress is higher than the interparticle repulsive force, the particles agglomerate and viscosity increases. In Figure 7c–e, we can see that after 50 cycles, STE obtains a smoother negative electrode surface, while the negative electrode surface of the aqueous electrolyte shows many flower-like dendrites, indicating that STE can alleviate the generation of dendrites, which plays a positive role in improving the performance of the battery. In Figure 7f,h, it can be seen that the prepared STE has a more constant voltage distribution, smaller overpotential, and better cycling performance than the conventional aqueous electrolytes over a period of 200 h. As shown in Figure 7i, during the charging and discharging process, when external stress is imposed, STE exhibits solid-like properties with increased mechanical strength to avoid short circuits; when the external stress is removed, it reverts to the liquid state to maintain the basic electrochemical properties. All of the above proves that the STE

can alleviate zinc dendrite growth, stabilize the electrolyte–electrode interface, and enhance the electrochemical properties as well as provide fine impact resistance. Lithium–oxygen batteries suffer from problems such as deterioration or volatilization of the liquid electrolyte because their open structure allows oxygen to enter and exit. The involvement of oxygen will accelerate the deterioration of the battery performance [64]. Zheng et al. [65] developed NNFQSE, an electrolyte with a shear thickening effect that maintains the ultra-long life of lithium–oxygen batteries. As shown in Figure 7j–l, the ionic dipole interaction between the polar-CFx group of PVDF-HFP and the sulfate group of SiO_2 gives NNFQSE a honeycomb morphology and surface channels with a depth of up to 60.7 nm, which allows faster movement of Li^+ and accommodates more liquid electrolyte, which can be confirmed by Figure 7o. After 360 h, the electrolyte retention rate remains 90.5%, significantly higher than the other two electrolytes. As shown in Figure 7m,n, the structural morphology and the vertical channels of the STE remain well after 5000 h of cycling. The minimum impedance of EIS curves for the full cells at room temperature also verifies the ability of NNFQSE to improve the ion transfer rate. As can be seen in Figure 7q,r, COMSOL simulations show that under the same external conditions, the dendrites of NNFQSE increase only from 3.5 μm to 3.9 μm within 300 s. However, the dendrites without NNFQSE increase from 3.5 μm to 7.8 μm, showing an increase rate of approximately 220%. This shows that the growth of lithium dendrites can be reduced and thus the life cycle of the battery can be improved. NNFQSE exhibits shear thinning to eliminate the strain under long-term Li plating/stripping and shear thickening to restrain the dendrites' growth. STEs are widely used in batteries and have a promising future. It is essential to restrain dendrite growth to prolong cell life; however, the stability of cell cycling and flame retardancy are also of concern.
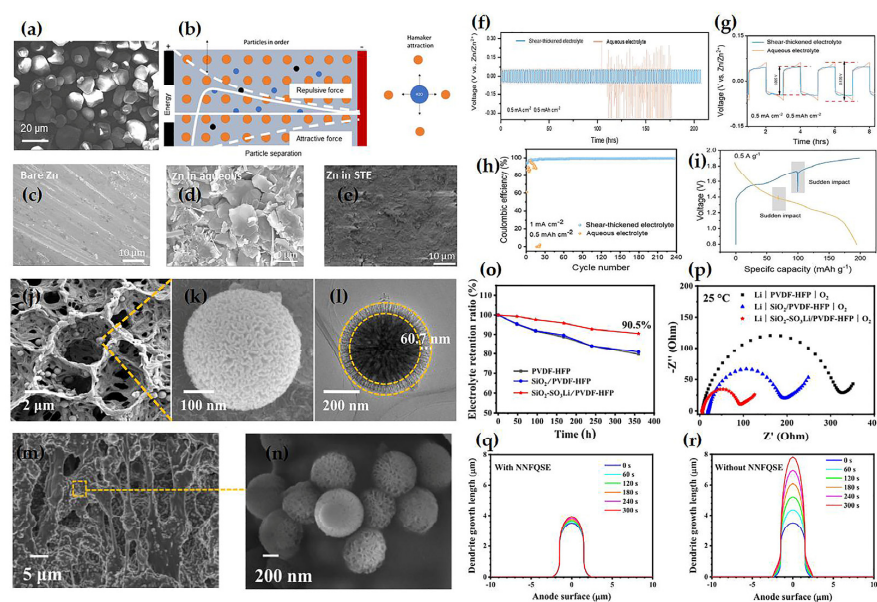
Figure 7. Performance of shear thickening electrolytes in zinc-ion and lithium–oxygen batteries: (**a**) SEM image of cornstarch particles. (**b**) schematic diagram of Hamaker attraction in the suspension. SEM images of (**c**) bare Zn negative electrode before cycling, (**d**) Zn negative electrode in aqueous electrolyte after cycling and (**e**) Zn negative electrode in cornstarch suspension electrolyte after cycling. (**f**) Zn plating and stripping with aqueous electrolyte and STE at a capacity of 0.5 mA h cm^{-2} and current density of 0.5 mA cm^{-2}. (**g**) Enlarged diagram of f. (**h**) Coulombic efficiency in Cu//Zn at 1 mA cm^{-2}. (**i**) Galvanostatic cycling test for Zn|STE|MnO_2 under sudden impact at 0.5 A g^{-1} [58]. Copyright 2022, Royal Society of Chemistry. (**j–l**) SEM images of SiO_2-SO_3Li/PVDF-HFP quasi-solid electrolyte and TEM image of the SiO_2-SO_3Li particles. (**m,n**) cross-section SEM images of NNFQSE SiO_2-SO_3Li/PVDF-HFP before and after 5000 h cycling. (**o**) electrolyte retention ratio. (**p**) EIS curves for the full cells. COMSOL simulations of lithium dendrite growth within 300 s (**q**) with NNFQSE or (**r**) without NNFQSE [65]. Copyright 2023, Nature.

4. Conclusions

This paper presents a review of shear thickening electrolytes based on a liquid–solid conversion mechanism, including a description of the principle, battery performance testing, and practical applications. First, we learned that as a non-Newtonian fluid, the normal state of shear thickening electrolyte is the slightly viscous liquid state. Once the shear thickening electrolyte is subjected to external shock, the shear storage modulus and other rheological properties increased significantly, and once the impact loading of the external force was off, the system reverted to the original liquid state. It has been found that the ST effect is related to the size of the dispersed particles, mass fraction, dispersion medium, ambient temperature, etc. The ST effect in the electrolyte system plays a protective role for the battery system when it subjects it to external mechanical abuse. In addition, electrochemical and ballistic tests show that the shear thickening electrolyte is capable of converting between liquid and solid phases, which is beneficial to the battery system. This makes the shear thickening electrolyte a combination of the advantages of both solid and liquid electrolytes, with a stable electrochemical performance similar to liquid electrolytes and good safety characteristics similar to solid electrolytes, providing a reference for the future development of high-safety performance batteries. The shear thickening electrolyte relies on the interaction between the fillers and solvent molecules, and the formation of nanoparticle clusters under drastic mechanical abuse conditions. In addition to its impact resistance and ionic conductivity, the performance of the shear thickening electrolyte is evaluated by the ability to form the quasi-state electrolyte. Furthermore, this type of rate-sensitive electrolyte is not limited to lithium-ion batteries but is also suitable for usage in sodium-ion and potassium-ion batteries, showing the potential for a wide range of application.

5. Perspectives

The development of shear thickening electrolytes is still in its early stages and faces a range of challenges in the practical application of high-performance lithium-ion batteries. Key criteria to assess the performance of this type of electrolyte include impact resistance, ionic conductivity, and interfacial compatibility with electrodes. In particular, the electrochemical properties of shear thickening electrolytes and the description of the mechanisms involved in the shear thickening process are still unsolved. The filler is added continuously until the shear thickening effect can occur. The specific amount of filler needs to be explored for different systems, where the structure of the filler and the composition of the electrolyte are also among the influencing factors. In addition, after adding the fillers, there is a slight decrease in energy density and specific energy, where the amount of decrease depends on the amount of filler added. Therefore, how to achieve the shear thickening effect with fewer additives while reducing the energy density loss is a direction to be explored in the future. As a new type of smart electrolytes, shear thickening electrolytes are expected to be widely applied in commercial full batteries, flexible wearable devices, etc. As shown in Figure 8, the application of shear thickening electrolytes to battery systems will provide valuable insights for future research on safety batteries. It is expected that future research will explore more effective strategies to improve the performance of shear thickening electrolyte batteries. With the solution of a series of key issues, the industrialization and wide application of shear thickening electrolyte batteries will become a reality.
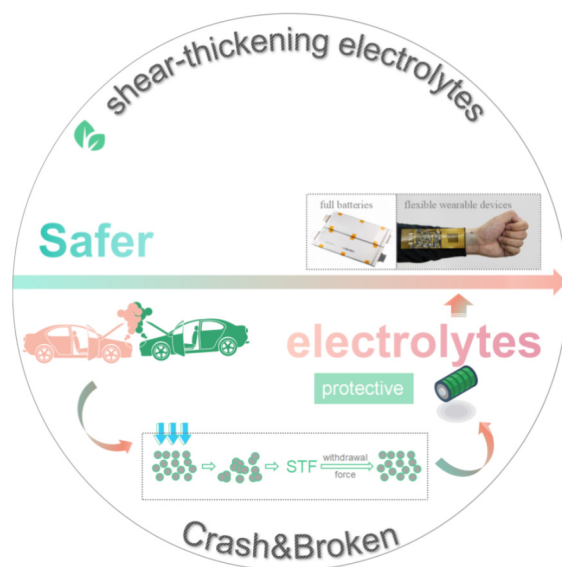
Figure 8. Summary and outlook chart in liquid–solid reversible shear thickening electrolytes.

References

1. Kalhoff, J.; Eshetu, G.G.; Bresser, D.; Passerini, S. Safer Electrolytes for Lithium-Ion Batteries: State of the Art and Perspectives. *ChemSusChem* **2015**, *8*, 2154–2175. [CrossRef] [PubMed]
2. Balakrishnan, P.G.; Ramesh, R.; Prem Kumar, T. Safety Mechanisms in Lithium-Ion Batteries. *J. Power Sources* **2006**, *155*, 401–414. [CrossRef]
3. Ming, J.; Cao, Z.; Wu, Y.; Wahyudi, W.; Wang, W.; Guo, X.; Cavallo, L.; Hwang, J.-Y.; Shamim, A.; Li, L.-J.; et al. New Insight on the Role of Electrolyte Additives in Rechargeable Lithium Ion Batteries. *ACS Energy Lett.* **2019**, *4*, 2613–2622. [CrossRef]
4. Liu, K.; Liu, Y.; Lin, D.; Pei, A.; Cui, Y. Materials for Lithium-Ion Battery Safety. *Sci. Adv.* **2018**, *4*, eaas9820. [CrossRef] [PubMed]
5. Ouyang, D.; Chen, M.; Huang, Q.; Weng, J.; Wang, Z.; Wang, J. A Review on the Thermal Hazards of the Lithium-Ion Battery and the Corresponding Countermeasures. *Appl. Sci.* **2019**, *9*, 2483. [CrossRef]
6. Arora, S.; Shen, W.; Kapoor, A. Review of Mechanical Design and Strategic Placement Technique of a Robust Battery Pack for Electric Vehicles. *Renew. Sustain. Energy Rev.* **2016**, *60*, 1319–1331. [CrossRef]
7. Zhang, L.; Huang, Y.; Fan, H.; Wang, H. Flame-Retardant Electrolyte Solution for Dual-Ion Batteries. *ACS Appl. Energy Mater.* **2019**, *2*, 1363–1370. [CrossRef]
8. Deng, K.; Zeng, Q.; Wang, D.; Liu, Z.; Wang, G.; Qiu, Z.; Zhang, Y.; Xiao, M.; Meng, Y. Nonflammable Organic Electrolytes for High-Safety Lithium-Ion Batteries. *Energy Storage Mater.* **2020**, *32*, 425–447. [CrossRef]
9. Hyung, Y.E.; Vissers, D.R.; Amine, K. Flame-Retardant Additives for Lithium-Ion Batteries. *J. Power Sources* **2003**, *119*, 383–387. [CrossRef]
10. Xiang, H.; Jin, Q.; Chen, C.H.; Ge, X.; Guo, S.; Sun, J. Dimethyl Methylphosphonate-Based Nonflammable Electrolyte and High Safety Lithium-Ion Batteries. *J. Power Sources* **2007**, *174*, 335–341. [CrossRef]

11. Kizilel, R.; Sabbah, R.; Selman, J.R.; Al-Hallaj, S. An Alternative Cooling System to Enhance the Safety of Li-Ion Battery Packs. *J. Power Sources* **2009**, *194*, 1105–1112. [CrossRef]

12. Kizilel, R.; Lateef, A.; Sabbah, R.; Farid, M.; Selman, J.; Al-Hallaj, S. Passive Control of Temperature Excursion and Uniformity in High-Energy Li-Ion Battery Packs at High Current and Ambient Temperature. *J. Power Sources* **2008**, *183*, 370–375. [CrossRef]

13. MacFarlane, D.R.; Forsyth, M.; Howlett, P.C.; Kar, M.; Passerini, S.; Pringle, J.M.; Ohno, H.; Watanabe, M.; Yan, F.; Zheng, W.; et al. Ionic Liquids and Their Solid-State Analogues as Materials for Energy Generation and Storage. *Nat. Rev. Mater.* **2016**, *1*, 15005. [CrossRef]

14. Shu, K.; Wang, C.; Li, W.; Bussell, T.; Ding, J. Electrolytes with Reversible Switch between Liquid and Solid Phases. *Curr. Opin. Electrochem.* **2020**, *21*, 297–302. [CrossRef]

15. Veith, G.M.; Armstrong, B.L.; Wang, H.; Kalnaus, S.; Tenhaeff, W.E.; Patterson, M.L. Shear Thickening Electrolytes for High Impact Resistant Batteries. *ACS Energy Lett.* **2017**, *2*, 2084–2088. [CrossRef]

16. Zhang, S.S. A Review on Electrolyte Additives for Lithium-Ion Batteries. *J. Power Sources* **2006**, *162*, 1379–1394. [CrossRef]

17. Haregewoin, A.M.; Wotango, A.S.; Hwang, B.-J. Electrolyte Additives for Lithium Ion Battery Electrodes: Progress and Perspectives. *Energy Environ. Sci.* **2016**, *9*, 1955–1988. [CrossRef]

18. Barnes, H. Shear-Thickening ("Dilatancy") in Suspensions of Nonaggregating Solid Particles Dispersed in Newtonian Liquids. *J. Rheol.* **1989**, *33*, 329–366. [CrossRef]

19. Wyart, M.; Cates, M.E. Discontinuous Shear Thickening without Inertia in Dense Non-Brownian Suspensions. *Phys. Rev. Lett.* **2014**, *112*, 098302. [CrossRef]

20. Bag, N.; Bhattacharyya, S. Electroosmotic Flow of a Non-Newtonian Fluid in a Microchannel with Heterogeneous Surface Potential. *J. Non-Newton. Fluid Mech.* **2018**, *259*, 48–60. [CrossRef]

21. Wagner, N.J.; Brady, J.F. Shear Thickening in Colloidal Dispersions. *Phys. Today* **2009**, *62*, 27–32. [CrossRef]

22. Olsson, P.; Teitel, S. Critical Scaling of Shear Viscosity at the Jamming Transition. *Phys. Rev. Lett.* **2007**, *99*, 178001. [CrossRef] [PubMed]

23. Liu, K.; Cheng, C.-F.; Zhou, L.; Zou, F.; Liang, W.; Wang, M.; Zhu, Y. A Shear Thickening Fluid Based Impact Resistant Electrolyte for Safe Li-Ion Batteries. *J. Power Sources* **2019**, *423*, 297–304. [CrossRef]

24. Ye, Y.; Xiao, H.; Reaves, K.; McCulloch, B.; Mike, J.F.; Lutkenhaus, J.L. Effect of Nanorod Aspect Ratio on Shear Thickening Electrolytes for Safety-Enhanced Batteries. *ACS Appl. Nano Mater.* **2018**, *1*, 2774–2784. [CrossRef]

25. Raghavan, S.R.; Walls, H.J.; Khan, S.A. Rheology of Silica Dispersions in Organic Liquids: New Evidence for Solvation Forces Dictated by Hydrogen Bonding. *Langmuir* **2000**, *16*, 7920–7930. [CrossRef]

26. Li, X.; Cao, H.; Gao, S.; Pan, F.; Weng, L.; Song, S.; Huang, Y. Preparation of Body Armour Material of Kevlar Fabric Treated with Colloidal Silica Nanocomposite. *Plast. Rubber Compos.* **2008**, *37*, 223–226. [CrossRef]

27. Zhang, X.; Li, W.; Gong, X. The Rheology of Shear Thickening Fluid (Stf) and the Dynamic Performance of an Stf-Filled Damper. *Smart Mater. Struct.* **2008**, *17*, 035027. [CrossRef]

28. Gürgen, S.; Kuşhan, M.C.; Li, W. Shear Thickening Fluids in Protective Applications: A Review. *Prog. Polym. Sci.* **2017**, *75*, 48–72. [CrossRef]

29. Zhang, J.; Wang, Y.; Zhou, J.; Wu, J.; Liu, S.; Sang, M.; Liu, B.; Pan, Y.; Gong, X. Multi-Functional Stf-Based Yarn for Human Protection and Wearable Systems. *Chem. Eng. J.* **2023**, *453*, 139869. [CrossRef]

30. Liu, B.; Liu, Q.; Pan, Y.; Zhou, J.; Zhang, J.; Liu, S.; Fan, Z.; Deng, H.; Hu, Y.; Gong, X. An Impact-Resistant and Flame-Retardant Cnts/Stf/Kevlar Composite with Conductive Property for Safe Wearable Design. *Compos. Part A Appl. Sci. Manuf.* **2023**, *168*, 107489. [CrossRef]

31. Zheng, Y.; Yin, R.; Zhao, Y.; Liu, H.; Zhang, D.; Shi, X.; Zhang, B.; Liu, C.; Shen, C. Conductive Mxene/Cotton Fabric Based Pressure Sensor with Both High Sensitivity and Wide Sensing Range for Human Motion Detection and E-Skin. *Chem. Eng. J.* **2021**, *420*, 127720. [CrossRef]

32. Zarei, M.; Aalaie, J. Application of Shear Thickening Fluids in Material Development. *J. Mater. Res. Technol.* **2020**, *9*, 10411–10433. [CrossRef]

33. Zhang, X.; Li, W.; Gong, X. Study on Magnetorheological Shear Thickening Fluid. *Smart Mater. Struct.* **2008**, *17*, 015051. [CrossRef]

34. Williams, T.H.; Day, J.; Pickard, S. Surgical and Medical Garments and Materials Incorporating Shear Thickening Fluids. U.S. Patent 12/440,086, 27 April 2009.

35. Liu, B.; Shelley, M.; Zhang, J. Focused Force Transmission through an Aqueous Suspension of Granules. *Phys. Rev. Lett.* **2010**, *105*, 188301. [CrossRef]

36. Wu, Y.; Wang, S.; Sang, M.; Shu, Q.; Zhang, J.; Xuan, S.; Gong, X. A Safeguarding and High Temperature Tolerant Organogel Electrolyte for Flexible Solid-State Supercapacitors. *J. Power Sources* **2021**, *505*, 230083. [CrossRef]

37. Zhang, P.; Wang, J.; Sheng, W.; Wang, F.; Zhang, J.; Zhu, F.; Zhuang, X.; Jordan, R.; Schmidt, O.G.; Feng, X. Thermoswitchable on-Chip Microsupercapacitors: One Potential Self-Protection Solution for Electronic Devices. *Energy Environ. Sci.* **2018**, *11*, 1717–1722. [CrossRef]

38. Zhu, M.; Meng, W.; Huang, Y.; Zhi, C. Proton-Insertion-Enhanced Pseudocapacitance Based on the Assembly Structure of Tungsten Oxide. *ACS Appl. Mater. Interfaces* **2014**, *6*, 18901–18910. [CrossRef]

39. Wang, X.; Wang, P.; Jiang, Y.; Su, Q.; Zheng, J. Facile Surface Modification of Silica Nanoparticles with a Combination of Noncovalent and Covalent Methods for Composites Application. *Compos. Sci. Technol.* **2014**, *104*, 1–8. [CrossRef]

40. Wang, X.; Wang, L.; Su, Q.; Zheng, J. Use of Unmodified Sio2 as Nanofiller to Improve Mechanical Properties of Polymer-Based Nanocomposites. *Compos. Sci. Technol.* **2013**, *89*, 52–60. [CrossRef]
41. Tripathi, A.M.; Su, W.-N.; Hwang, B.J. In Situ Analytical Techniques for Battery Interface Analysis. *Chem. Soc. Rev.* **2018**, *47*, 736–851. [CrossRef]
42. Famprikis, T.; Canepa, P.; Dawson, J.A.; Islam, M.S.; Masquelier, C. Fundamentals of Inorganic Solid-State Electrolytes for Batteries. *Nat. Mater.* **2019**, *18*, 1278–1291. [CrossRef] [PubMed]
43. Isaac, J.A.; Devaux, D.; Bouchet, R. Dense Inorganic Electrolyte Particles as a Lever to Promote Composite Electrolyte Conductivity. *Nat. Mater.* **2022**, *21*, 1412–1418. [CrossRef] [PubMed]
44. Xu, X.; Wang, Y.; Yi, Q.; Wang, X.; Camacho, R.A.P.; Kungl, H.; Eichel, R.A.; Lu, L.; Zhang, H. Ion Conduction in Composite Polymer Electrolytes: Potential Electrolytes for Sodium-Ion Batteries. *ChemSusChem* **2023**, *16*, e202202152. [CrossRef] [PubMed]
45. Shen, C.; Wang, J.; Tang, Z.; Wang, H.; Lian, H.; Zhang, J.; Cao, C.-n. Physicochemical Properties of Poly (Ethylene Oxide)-Based Composite Polymer Electrolytes with a Silane-Modified Mesoporous Silica Sba-15. *Electrochim. Acta* **2009**, *54*, 3490–3494. [CrossRef]
46. Chen-Yang, Y.; Wang, Y.; Chen, Y.; Li, Y.; Chen, H.; Chiu, H. Influence of Silica Aerogel on the Properties of Polyethylene Oxide-Based Nanocomposite Polymer Electrolytes for Lithium Battery. *J. Power Sources* **2008**, *182*, 340–348. [CrossRef]
47. Croce, F.; Appetecchi, G.; Persi, L.; Scrosati, B. Nanocomposite Polymer Electrolytes for Lithium Batteries. *Nature* **1998**, *394*, 456–458. [CrossRef]
48. Jayanthi, S.; Shenbagavalli, S.; Muthuvinayagam, M.; Sundaresan, B. Effect of Nano Tio2 on the Thransport, Structural and Thermal Properties of Pema-Nai Solid Polymer Electrolytes for Energy Storage Devices. *Mater. Sci. Eng. B* **2022**, *285*, 115942. [CrossRef]
49. Volfkovich, Y.M.; Rychagov, A.Y.; Sosenkin, V.E.; Baskakov, S.A.; Kabachkov, E.N.; Shulga, Y.M. Supercapacitor Properties of Rgo-Tio2 Nanocomposite in Two-Component Acidic Electrolyte. *Materials* **2022**, *15*, 7856. [CrossRef]
50. Bronstein, L.M.; Karlinsey, R.L.; Ritter, K.; Joo, C.G.; Stein, B.; Zwanziger, J.W. Design of Organic–Inorganic Solid Polymer Electrolytes: Synthesis, Structure, and Properties. *J. Mater. Chem.* **2004**, *14*, 1812–1820. [CrossRef]
51. Kumar, B.; Fellner, J. Polymer–Ceramic Composite Protonic Conductors. *J. Power Sources* **2003**, *123*, 132–136. [CrossRef]
52. Charradi, K.; Ahmed, Z.; Aranda, P.; Chtourou, R. Silica/Montmorillonite Nanoarchitectures and Layered Double Hydroxide-Speek Based Composite Membranes for Fuel Cells Applications. *Appl. Clay Sci.* **2019**, *174*, 77–85. [CrossRef]
53. Ding, J.; Tian, T.; Meng, Q.; Guo, Z.; Li, W.; Zhang, P.; Ciacchi, F.T.; Huang, J.; Yang, W. Smart Multifunctional Fluids for Lithium Ion Batteries: Enhanced Rate Performance and Intrinsic Mechanical Protection. *Sci. Rep.* **2013**, *3*, 2485. [CrossRef]
54. Xiaoxia, C.; Kai, L.; Baoguo, W. Research on High-Safety Electrolytes and Their Application in Lithium-Ion Batteries. *Energy Storage Sci. Technol.* **2020**, *9*, 583–592. [CrossRef]
55. Yu, O.; Wenhui, H.; Kai, L. Research Progress of Smart Safety Electrolytes in Lithium-Ion Batteries. *Energy Storage Sci. Technol.* **2022**, *11*, 1772. [CrossRef]
56. Heo, Y.; Larson, R.G. The Scaling of Zero-Shear Viscosities of Semidilute Polymer Solutions with Concentration. *J. Rheol.* **2005**, *49*, 1117–1128. [CrossRef]
57. Srivastava, A.; Majumdar, A.; Butola, B.S. Improving the Impact Resistance of Textile Structures by Using Shear Thickening Fluids: A Review. *Crit. Rev. Solid State Mater. Sci.* **2012**, *37*, 115–129. [CrossRef]
58. Dong, H.; Hu, X.; He, G. A Shear-Thickening Colloidal Electrolyte for Aqueous Zinc-Ion Batteries with Resistance on Impact. *Nanoscale* **2022**, *14*, 14544–14551. [CrossRef]
59. Hamaker, H.C. The London—Van Der Waals Attraction between Spherical Particles. *Physica* **1937**, *4*, 1058–1072. [CrossRef]
60. Mewis, J.; Biebaut, G. Shear Thickening in Steady and Superposition Flows Effect of Particle Interaction Forces. *J. Rheol.* **2001**, *45*, 799–813. [CrossRef]
61. Liu, Z.; Yang, Y.; Liang, S.; Lu, B.; Zhou, J. Ph-Buffer Contained Electrolyte for Self-Adjusted Cathode-Free Zn–Mno2 Batteries with Coexistence of Dual Mechanisms. *Small Struct.* **2021**, *2*, 2100119. [CrossRef]
62. Yufit, V.; Tariq, F.; Eastwood, D.S.; Biton, M.; Wu, B.; Lee, P.D.; Brandon, N.P. Operando Visualization and Multi-Scale Tomography Studies of Dendrite Formation and Dissolution in Zinc Batteries. *Joule* **2019**, *3*, 485–502. [CrossRef]
63. Hsu, P.-C.; Seol, S.-K.; Lo, T.-N.; Liu, C.-J.; Wang, C.-L.; Lin, C.-S.; Hwu, Y.; Chen, C.; Chang, L.-W.; Je, J.; et al. Hydrogen Bubbles and the Growth Morphology of Ramified Zinc by Electrodeposition. *J. Electrochem. Soc.* **2008**, *155*, D400. [CrossRef]
64. Yi, J.; Guo, S.; He, P.; Zhou, H. Status and Prospects of Polymer Electrolytes for Solid-State Li–O_2 (Air) Batteries. *Energy Environ. Sci.* **2017**, *10*, 860–884. [CrossRef]
65. Zheng, G.; Yan, T.; Hong, Y.; Zhang, X.; Wu, J.; Liang, Z.; Cui, Z.; Du, L.; Song, H. A Non-Newtonian Fluid Quasi-Solid Electrolyte Designed for Long Life and High Safety Li-O_2 Batteries. *Nat. Commun.* **2023**, *14*, 2268. [CrossRef] [PubMed]