

离子液体基固态电解质的研究进展

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摘要

固态电池由于具备高的理论能量密度和高安全性, 被视为下一代储能技术重要发展方向。固态电解质是固态电池关键组成部分, 其性能直接影响了固态电池的发展。离子液体基固态电解质具备高的离子电导率、高的化学稳定性和良好的机械性能, 在固态电池领域具备巨大的潜力。本文重点论述了离子液体基固态电解质的研究进展, 对离子液体基固态电解质的类型进行归纳总结, 并详细介绍了其在固态锂电池、固态钠电池、固态超级电容器三种储能技术中的应用优势。最后, 对离子液体基固态电解质的研究方向进行了展望。

关键词

离子液体, 固态电解质, 固态锂电池, 固态钠电池, 固态超级电容器

Research Progress of Ionic Liquid Based Solid Electrolyte

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Abstract

Solid-state batteries are widely regarded as a key advancement in next-generation energy storage due to their high theoretical energy density and superior safety. A crucial component of these batteries is the solid-state electrolyte, which directly impacts their performance and development. Among various electrolyte options, ionic liquid-based solid-state electrolytes have gained significant attention due to their high ionic conductivity, excellent chemical stability, and robust mechanical

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properties. This paper reviews the research progress in the development of ionic liquid-based solid-state electrolytes, categorizes their different types, and examines their advantages in three major energy storage technologies: solid-state lithium batteries, solid-state sodium batteries, and solid-state supercapacitors. Finally, future research directions and challenges in this field are discussed.

Keywords

Ionic Liquid, Solid Electrolyte, Solid Lithium Battery, Solid Sodium Battery, Solid Supercapacitor

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1. 引言

随着科技的变革和经济的发展,新能源行业迫切需要开发能量密度更高、使用寿命更长的储能技术[1]。二次电池是一种高效、清洁、可多次循环的电化学储能技术,已被广泛应用于电动汽车、便携电子等领域[2][3]。电解质是二次电池最重要的组件之一,在充放电过程中担任离子迁移的媒介,是研究者关注的热点。固态电解质具备较宽的电化学窗口和良好的机械性能,能够实现高电压正极材料和锂负极材料在固态电池中的应用,从根本上解决了传统液态电解液易泄露、易燃烧等安全问题,被视为下一代储能技术的重要研究方面[4][5]。近年来,研究者对固态电解质的制备开展了大量的研究,主要分为聚合物有机固态电解质、无机固态电解质和复合固态电解质三种[6][7]。然而,固态电解质的性能并不理想,离子电导率低、机械性能差等问题限制其进一步的发展,固态电解质的研发也成为制约固态电池技术发展的主要问题[7]。

离子液体是一种由阴阳离子组成的室温有机熔盐,具备高电导率、高热稳定性高、不易燃烧及电化学稳定性好等优势,被广泛应用于固态电解质的研究[8]-[10]。一方面,离子液体具有独特的物理化学性质,在室温下为液态且不易挥发,这使得基于离子液体的固态电解质在高温环境下也能保持稳定,极大地降低了因挥发导致的电池性能衰退和安全隐患[11][12]。另一方面,离子液体的高离子电导性确保了在电池充放电过程中锂离子能够快速、高效地传输,有助于提高电池的倍率性能和充放电效率[13]。另外,良好的热稳定性使离子液体能够在较宽的温度范围内保持稳定的物理化学性质,适应不同的应用环境,拓宽了电池的应用场景[14]。本文概述了几种不同类型离子液体基固态电解质的研究进展,并详细介绍了离子液体基固态电解质在固态锂电池、固态钠电池、固态超级电容器三种储能技术中的应用,最后对离子液体固态电解质发展方向进行了展望。

2. 离子液体基固态电解质的研究进展

将离子液体掺杂到聚合物基体中,如聚环氧乙烷(PEO)、聚偏氟乙烯(PVDF-HFP)、聚乙二醇(PEG)等,可以提高聚合物电解质的柔性和离子传输能力,形成离子液体基聚合物固态电解质[15]-[17]。离子液体与聚合物基体的相互作用能够影响聚合物链段的运动能力[18]。例如,吡咯烷离子液体的阳离子能够与 PEO 链段上的氧原子形成强的氢键作用,这不仅降低了 PEO 的结晶度,提高了聚合物的柔韧性,还增加了锂离子的传输通道,从而提升了电解质的离子电导率[19]。同时,离子液体的高热稳定性也赋予了聚合物电解质在较宽温度范围内的稳定性,使其能够在高温环境下保持良好的性能。Liton Balo 等将 1-乙基-3-甲

咪唑双三氟甲烷磺酰亚胺盐(EMIMTFSI)离子液体引入 PEO 聚合物基底中制备出柔性凝胶聚合物电解质(GPE), 明显提高了电解质的室温离子电导率, 组装出电池具有良好的放电比容量[20]。韩国延世大学 Jong Hyeok Park 教授将吡咯烷离子液体与线性聚乙二醇共聚酯共聚得到离子液体基聚合物固态电解质(IPSE), 这种电解质具备较低的玻璃转换温度与较高的离子电导率, 电池表现出良好的倍率性能[21]。另外, 离子液体有利于帮助固态电解质在电极/电解质截面衍生界面保护层, 进而提高电池的循环稳定性[11]。Liang 等人[22]合成了一种新型的双功能离子液体, 通过开环聚合的方法形成双交联聚(离子液体)电解质(PIL-PEI)。这种 PIL-PEI 电解质具有良好的柔韧性、热稳定性和优异的电化学性能, 其室温电导率达到 $1.03 \times 10^{-3} \text{ S/cm}$ 。此外, PIL-PEI 的网络结构能够在锂金属上形成稳定的固体电解质界面层(SEI), 从而抑制锂枝晶生长。

将离子液体引入无机固态电解质体系中, 可以有效降低界面电阻, 增强界面稳定性[23] [24]。Maximilian Fichtner 等人在无机固态电解质 LLZO 与电极界面添加了少量 Li [TFSA] [C4C1pyrr] [FSA]离子液体, 改善了电极与固体电解质的接触, 显著降低了正极和负极界面的界面电阻[25]。Li [TFSA] [C4C1pyrr] [FSA]离子液体能够在 LLZO 表面形成富含有机成分的界面层, 这层界面层能够有效降低电极与电解质之间的界面电阻, 提高锂离子在界面处的传输效率。此外, 离子液体形成的界面层能够均匀化锂离子的沉积过程, 避免局部锂离子浓度过高而导致枝晶的形成, 进而抑制了锂枝晶的生长。最终, 引入离子液体后 Li/LLZO/LiFePO₄ 电池展示出良好的电化学性能, 放电比容量达到 145 mAh/g, 库伦效率超过 99%。此外, 将离子液体与聚合物固态电解质、无机固态电解质复合, 可以得到综合性能优良的离子液体基复合固态电解质。Zhengkun Xie 等人将 PEO、双三氟甲烷磺酰亚胺锂(LiTFSI)、四丁基磷 2-羟基吡啶(TBPHP)和 Li_{6.4}La₃Zr_{1.4}Ta_{0.6}O₁₂(LLZTO)原料复合, 制备了一种柔性离子液体基固态电解质[26]。其中, TBPHP 离子液体作为增塑剂, 有效调节了 PEO 的结晶度, 促进了锂离子的迁移, 进而提高了固态电解质的离子电导率($9.39 \times 10^{-4} \text{ S/cm}$), 扩展了固态电解质的电化学稳定窗口(>5 V)。

3. 离子液体聚合物固态电解质在储能技术中的应用

通过上述研究进展可以看出, 离子液体基固态电解质在材料性能方面取得了巨大的突破, 其高离子电导率、良好的化学稳定性和优异的机械性能为其在储能技术领域的应用奠定了坚实的基础。接下来, 将进一步探讨离子液体基固态电解质在不同储能技术中的实际应用情况, 分析其在提升储能器件性能方面所发挥的关键作用。

3.1. 离子液体固态电解质在固态锂电池中的应用

近年来, 离子液体聚合物固态电解质已成功应用于固态锂电池, 在提高固态电解质锂离子电导率的同时, 还可以促进锂均匀沉积/溶解, 使固态锂电池展现出优异的循环稳定性和倍率性能[15] [27]。Chen 等人[28]以 1-丁基-3-乙基咪唑双三氟甲磺酰亚胺盐(imidazolium IL)、乙烯基碳酸乙烯酯(VEC)、八氟己二醇二丙烯酸酯(OFHDDA)为单体, 结合双三氟甲磺酰亚胺锂盐(LiTFSI), 通过紫外光固化技术设计并合成了具有梯度性锂离子迁移数的聚合离子液体(PIL)电解质。通过优化单体比例, 成功将锂离子迁移数从 0.09 提升至 0.39。该电解质应用于 Li/NCM622 电池, 在 0.2 C 下 200 次循环后仍保持 84%的容量保持率。I. De Meatz 等人以聚二烯基二甲铵双(三氟甲磺酰基)亚胺聚合物离子液体(PIL)为聚合物主体, 掺入 PYR₁₄TFSI 离子液体和锂盐 LiTFSI, 形成一种新型离子液体聚合物固态电解质(PIL-LiTFSI-PYR₁₄TFSI) [29]。这种 PIL-LiTFSI-PYR₁₄TFSI 具有良好的化学稳定性、较好的室温离子电导率和较宽的电化学稳定窗口, 能够实现均匀锂沉积/溶解, 应用于 Li/LiFePO₄ 固态锂电池中放电比容量在 40℃达到 140 mAh/g。Kim 等人[30]将 LLZO 和 Li [C₄C₁pyrr] [TFSA]混合球磨, 得到离子液体基无机固态电解质, 其离子电导

率在室温下达到 0.4 mS/cm。而且, 离子液体的加入显著抑制锂枝晶的形成, 在 Li/LiFePO₄ 电池中循环 150 次后容量保持率高达 99%。Wu 等人将一种 MOF 材料(UiO-66)与离子液体复合, 得到了具有纳米结构的离子液体基固态电解质[31]。这种新型纳米结构的离子液体基固态电解质实现了锂离子在电极/电解质界面上的快速, 室温下离子电导率为 3.2×10^{-4} S/cm; 另外, 这种电解质使界面处形成稳定的固体导电层, 有效降低了电极/电解质界面, 使得锂镀/剥离稳定。基于这种离子液体固态电解质具有高放电比容量和优异的容量保留率, 0.2 C 放电比容量为 130 mAh/g, 循环 100 周后容量保持率接近 100%。

3.2. 离子液体固态电解质在固态钠电池中的应用

固态钠离子电池因其丰富的资源和低成本优势, 被视为锂离子电池的潜在替代方案[32]-[34]。采用离子液体固态电解质能够提升其离子导电性和电化学稳定性, 从而优化电池性能。Song 等人通过将 PEO、NaClO₄、SiO₂ 和 EMIFSI 原料混合, 制备了一种离子液体复合固态电解质, 用于固态钠电池[35]。这种离子液体复合固态电解质具备高的室温离子电导率(1.3×10^{-3} S cm⁻¹), 合适的力学性能, 的宽电压稳定窗口(4.2 V)和的高 Na⁺迁移数(0.61), 组装的固态钠电池可以在高温 60℃ 下循环 100 圈。Kumar 等人将钠盐三氟甲基磺酸钠(NaCF₃SO₃)和离子液体 1-乙基-3-甲基咪唑三氟甲磺酰盐(EMITf)引入 PVDF-HFP 基底中, 制备了一种基于固态钠电池的离子液体凝胶固态电解质[36]。这种离子液体凝胶固态电解质具有透明而紧凑的外观, 离子液体提供了离子传输通道, 促进钠离子迁移。最终, 该电解质在室温下表现出高的离子电导率(5.74×10^{-3} S/cm)和较高的 Na⁺迁移数(0.23)。Jinkwang Hwang 等人利用一种基于六氟磷酸酯的离子液体([DEME][PF₆])作为界面层, 采用 BASE/IL 结构组装了一种采用高压正极 Na₃V₂(PO₄)₂F₃ 的固态钠离子电池。[DEME][PF₆]的引入解决了固态电解质在高电压下氧化稳定性不足的问题, 并改善固态电解质与正极材料之间的界面接触, 显著提高了固态钠离子电池的氧化稳定性和界面接触, 从而大幅提升了钠电池的循环性能和能量密度[37]。该电池在 60℃ 下展现出 110 mAh/g 的可逆容量和 416 Wh/kg 的能量密度, 并在 1500 次循环后保持 107% 的容量保持率和 99.3% 的平均库仑效率。

3.3. 离子液体固态电解质在固态超级电容器中的应用

超级电容器因其高功率密度、长循环寿命和快速充放电能力, 在储能领域具有重要应用[38][39]。开发出具有优异电化学性能和热稳定性的固体电解质应用于超级电容器, 可以显著提高电容器的工作电压与能量密度[38][40]。离子液体基固态电解质可以提高离子电导率、提升电化学窗口, 在固态超级电容器中有着良好的应用前景。Marcilla 等人将聚二烯丙基二甲基铵双(三氟甲磺酰亚胺)与 1-甲基丁基吡咯双(三氟甲磺酰亚胺基)盐[PY₁₄][TFS]复合, 制备一种应用于固体超级电容器的离子液体基固态电解质, 具有优良的电化学性能[41]。这种离子液体基固态电解质具有高电化学稳定性, 使得固态超级电容器首次能够在高达 3.5 V 的最大电压下工作。Tu 等人将离子液体 1-丁基-3-甲基咪唑碘化物(BMIMI)作为增塑剂和氧化还原添加剂, 制备了一种具有氧化还原活性的聚乙烯醇(PVA)-Li₂SO₄-BMIMI 凝胶聚合物电解质(GPE), 并将其用于活性炭基柔性超级电容器的组装[42]。这种离子液体凝胶固态电解质组装的柔性超级电容器可以达到 29.3 Wh/kg 的高能量密度, 并具有良好的循环耐久性和优异的力学性能。Tang 等人[43]通过紫外光固化原位微相分离策略, 以 N-异丙基丙烯酰胺(NIPAM)和丙烯酸(AA)为单体、1-乙基-3-甲基咪唑硫酸乙酯(EMIES)为离子液体溶剂, 设计合成了微相分离结构离子凝胶电解质(PNIPAM-co-PAA)。该材料通过硬相(高结晶性 PNIPAM)与软相(非晶态 PAA-EMIES 复合相)的协同作用, 实现了 11.73 MPa 的断裂强度、473% 的拉伸应变及 28.09 MJ/m³ 的韧性, 同时具备 1.1 mS/cm 的离子电导率。基于此离子凝胶组装的全固态超级电容器, 在 400% 拉伸应变下展现出 228.1 mF/cm² 的高面积比电容和 5.82 MPa 的机械强度, 并在 5000 次折叠或 500 次 400% 拉伸循环后仍保持 90% 以上的容量保持率, 验证了其在柔性可穿戴电子

中的高可靠性和耐久性。

4. 总结与展望

开发具有高离子电导率、优良机械性能和良好化学/电化学稳定性的固态电解质是下一代储能技术的重要发展方向。离子液体可以显著改善固态电解质的离子电导率、增强固态电解质电化学稳定性、提高固态电解质的机械性能,因此,离子液体基固态电解质在新一代储能技术中具有广阔的应用前景。本文详细介绍了离子液体基固态电解质的研究进展及其在固态锂电池、固态钠电池与固态超级电容器中的应用。尽管离子液体基固态电解质的研发已取得重要进展,但仍面临诸多挑战。比如目前大多数离子液体基固态电解质的导电率在室温下仍较低,需要进一步优化材料设计。另外,离子液体工业化生产仍面临挑战,需开发低成本、高性能的离子液体基固态电解质材料。未来研究将致力于开发新型离子液体及其复合体系,通过合理的材料设计和优化,可以开发出兼具高安全性、高导电性和优异循环稳定性的固态电解质,为下一代储能技术的发展提供重要支撑。

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