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Zhengqi Wang et al., J Fundam Renewable Energy Appl 2018, Volume 8
DOI: 10.4172/2090-4541-C5-062

3rd International Conference on

Battery and Fuel Cell Technology

September 10-11, 2018 | London, UK

Selection of safe salt-in-carbonate electrolytes for lithium-ion batteries

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While proceeding to drive down costs and boosts capacity, lithium-ion batteries (LIBs) face challenges in safety issues, such as the storage reliability, the cell safety in terms of burning and thermal runaway and the work stability of the electrolyte. Our focus is therefore the prevention of these main risks associated with electrolytes. For preparation of suitable "salt-in-solvent" electrolyte mixtures, selected thermal stable conducting salts, e.g. lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), lithium tetrafluoroborate ($LiBF_4$) and lithium difluoro(oxalate)borate (LiDFOB) were dissolved in several high-boiled carbonates. The carbonates of different structures are available, e.g. cyclic ethylene carbonate (EC), propylene carbonate (PC), 1, 2-butylene carbonate (1, 2-BC) and fluoroethylene carbonate (FEC), and e.g. linear dibenzyl carbonate (DBC) and dipropyl carbonate (DPrC). Various possibilities inspired the interest to understand the relationship between molecular structures, composition of components and the properties of the electrolyte mixtures. Regarding the practical use, the solubility and chemical stability of salt-carbonate mixtures were investigated. We measured physicochemical properties (phase transition behaviour, density and viscosity) to draw a picture of interaction between ions and carbonate molecules. The temperature dependent viscosity η and ions conductivity κ of the mixtures could be well described by the empirical Vogel-Fulcher-Tammann (VFT) equation and correlated by the fractional Walden rule. The flow activation energy Ea was calculated and compared with each other regarding to Angell fragility concept. The electrochemical stability obtained by the cyclic voltammetry revealed the oxidative potentials of selected mixtures. The aluminum (Al) corrosion of mixtures with and without additives (e.g. lithium bis(oxalato) borate) was investigated by cell-cycling versus Li/Li+. Charge-discharge cycling performance of the electrolyte mixtures were investigated by full-cell tests using $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ and graphite as electrode materials. Due to the electrochemical damage or the Al corrosion, electrolytes might show unstable cycling results (e.g. irreversible loss of discharge capacity (mAh/g), capacity conservation problems). These phenomena could be significantly improved or avoided by utilizing additive, which forms a passivation film on the electrode surfaces. Besides, the flash point (>140°C) and the burning tests for the selected mixtures revealed safety improvement regarding to the practical aspects such as the vapour-leakage of the boiled electrolytes. In summary, the electrochemical measurements showed various properties influenced by the structure/composition and indicated that novel electrolyte mixtures could be regarded as stable and better performance candidates for basic electrolytes in lithium-ion batteries for safety improvement compared to state-of-the-art low boiling electrolyte mixtures.

Recent Publications:

1. A Hofmann et al., (2016) Investigation of binary mixtures containing 1-Ethyl-3-methylimidazolium Bis(trifluoromethanesulfonyl)azanide and ethylene carbonate. Journal of Chemical & Engineering Data 61:114-123.
2. A Hofmann et al., (2014) Anodic aluminum dissolution of LiTFSA containing electrolytes for Li-Ion-batteries. Electrochimica Acta 116:388-395.

Biography

Zhengqi Wang received his Bachelor degree in Material Science and Technology from Tongji University, China and a Master degree in Material Science and Material Technology from Karlsruhe Institute of Technology (KIT). From 2016 as a PhD student, he worked at Institute for Applied Materials (IAM-WK) of KIT. His research focus is the safety improvement of lithium ion battery by combination of innovative strategies with the different parts in batteries such as electrolytes, electrode materials. The various physiochemical characterizations, electrochemical measurements and cell tests will be used for analysis of the properties and the performances of lithium ion batteries.

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