ADVANCED ENERGY MATERIALS

# Fluorine-Free Noble Salt Anion for High-Performance All-Solid-State Lithium–Sulfur Batteries

*Heng Zhang, Xabier Judez, Alexander Santiago, Maria Martinez-Ibañez, Miguel Ángel Muñoz-Márquez, Javier Carrasco, Chunmei Li,* Gebrekidan Gebresilassie Eshetu,* and Michel Armand**

Dr. H. Zhang, X. Judez, Dr. A. Santiago, Dr. M. Martinez-Ibañez,
Dr. M. Á. Muñoz-Márquez, Dr. J. Carrasco, Dr. C. Li, Prof. M. Armand
Electrical Energy Storage Department
CIC Energigune
Parque Tecnológico de Álava
Albert Einstein 48, 01510 Miñano, Álava, Spain
E-mail: cli@cicenergigune.com; marmand@cicenergigune.com

Dr. G. G. Eshetu
Institute for Power Electronics and Electrical Drives (ISEA)
RWTH Aachen University
Jägerstraße 17/19, 52066 Aachen, Germany
E-mail: Gebrekidan.Eshetu@isea.rwth-aachen.de

Dr. G. G. Eshetu
Department of Chemistry
College of Natural and Computational Sciences
Mekelle University
P.O. Box-231, Mekelle, Ethiopia

First published: 27 May 2019 | https://doi.org/10.1002/aenm.201900763

## Abstract

Amongst post-Li-ion battery technologies, lithium–sulfur (Li–S) batteries have captured an immense interest as one of the most appealing devices from both the industrial and academia sectors. The replacement of conventional liquid electrolytes with solid polymer electrolytes (SPEs) enables not only a safer use of Li metal (Li°) anodes but also a flexible design in the shape of Li–S batteries. However, the practical implementation of SPEs-based all-solid-state Li–S batteries (ASSLSBs) is largely hindered by the shuttling effect of the polysulfide intermediates and the formation of dendritic Li° during the battery operation. Herein, a fluorine-free noble salt anion, tricyanomethanide [$C(CN)_3^-$, $TCM^-$], is proposed as a Li-ion conducting salt for ASSLSBs. Compared to the widely used perfluorinated anions {e.g., bis(trifluoromethanesulfonyl)imide anion, $[N(SO_2CF_3)_2)]^-$, $TFSI^-$}, the LiTCM-based electrolytes show decent ionic conductivity, good thermal stability, and sufficient anodic stability suiting the cell chemistry of ASSLSBs. In particular, the fluorine-free solid electrolyte interphase layer originating from the decomposition of LiTCM exhibits a good mechanical integrity and Li-ion conductivity, which allows the LiTCM-based Li–S cells to be cycled with good rate capability and Coulombic efficiency. The LiTCM-based electrolytes are believed to be the most promising candidates for building cost-effective and high energy density ASSLSBs in the near future.