**Identifying the components of the solid–electrolyte interphase in Li-ion Batteries**

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This document presents the raw data file from the solid- state NMR experiments and CASTEP calculations.

**1. Raw files from NMR experiments**

**Supplementary Figure 6.** 1D solid state NMR spectra.(a)1H (850 MHz) and (b) 7Li (330 MHz) one pulse MAS NMR spectra of LMC, LEMC and LEDC·2DMSO. (c) LMC and (d) LEMC 1H (850 MHz) MAS (60 kHz) NMR spectra, together with the stick spectra corresponding to the GIPAW calculated chemical shifts for the geometry optimized (CASTEP) crystal structure models. \*: impurity (hydrolysis) due to sample preparation.

1. 1H (850 MHz) one pulse MAS (60 kHz): **SI-Figure6****1H**LMC [1] (recorded on 06/04/18) , LEMC [1] (recorded on 05/04/18) and LEDC.2DMSO [1] (recorded on 05/04/18)
2. 7Li (330 MHz) one pulse MAS (60 kHz): **SI-Figure6****7Li**LMC [2] (recorded on 06/04/18), LEMC [2] (recorded on 05/04/18), and LEDC.2DMSO [2] (recorded on 05/04/18)

**Supplementary Figure 7.** (a1, b1 and c1) 2D 1H (850 MHz) (DQ)-1H (SQ) MAS (60 kHz, with 1 τR of BABA recoupling), and (a2, b2 and c2) 2D 7Li-1H (850 MHz) HMQC MAS (60 kHz, τRCPL= 400 μs) NMR spectra of (a) LMC, (b) LEMC and (c) LEDC·2DMSO. For the 2D spectra, skyline projections are presented; in addition, at the top a 1D one-pulse 1H MAS NMR spectrum (dashed) is presented. The base contour levels are at 4 % and 14 % of the maximum peak intensity in (a1) and (a2) for LMC, 38 % and 44 % in (b1) and (b2) for LEMC, 22% and 20% for (c1) and (c2) for LEDC·2DMSO, respectively.

1. 1H (850 MHz) DQ spectrum : **SI-Figure7** – **1H DQ**  LMC [4] (recorded on 06/04/18), LEMC [4] (recorded on 05/04/17), LEDC.2DMSO [4] ( recorded on 05/04/18)
2. 2D 7Li-1H (850 MHz) HMQC MAS : **SI-Figure7****7L1H HMQC**- LMC [11] ( recorded on 06/04/18), LEMC [5] ( recorded on 05/04/18), LEDC.2DMSO [5] (recorded on 05/05/18)

**Supplementary Figure 25**. 1D solid state NMR spectra.(a)1H (850 MHz) and (b) 7Li (330 MHz) one-pulse MAS NMR spectra of SEI layers @ graphite powders. Very broad peaks were observed, likely due to paramagnetism.

1. 1H (850 MHZ) MAS (36 kHz) : **SI-Figure25SEI layer@graphite1H[1] (recorded on 29/05/18)**
2. 7Li (330 MHz) MAS (36 kHz): **SI-Figure25SEI layer@graphite7Li [4] (recorded on 29/05/18)**

**2. Calculations**

**For the CASTEP calculations of LEMC and LMC, the initial cif file and geometry optimized cif file and the magres file for the full crystal structure**

**LEMC**

LEMC.cif- (determined from the single crystal x-ray diffraction)

LEMC-out.cif (geometry optimized full crystal structure)

LEMC\_NMR.magres (contains the chemical shieldings)

**LMC1**

LMC1.cif

LMC1-out.cif

LMC1\_NMR.magres

**LMC2**

LMC2.cif

LMC2-out.cif

LMC2\_NMR.magres