Creep deformation of lithium foil at moderate pressures (< 2 MPa) and temperatures (30-50°C)

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Abstract text:

Lithium metal is receiving significant research attention as a potential next generation negative electrode material in lithium ion batteries because it can offer a ~50% increase in cell energy density compared to conventional graphite electrodes. There are, however, a number of mechanical challenges using lithium metal electrodes. Mechanical stack pressure is required to ensure good contact between solid-state electrodes and electrolytes. Moderate to high stack pressures (e.g.< 10 MPa) can also supress void formation during fast charging and discharging.[1] Creep, the plastic deformation of a material under constant pressure, improves battery performance by preventing and filling voids, but can also lead to decreased performance if lithium is unable to sustain the required stack pressure, and potentially cell shorting if lithium is e.g. extruded beyond the current collectors.

Creep deformation rates are highly dependent on homologous temperature and pressure. Lithium metal has a relatively low melting point (181 °C) / high homologous temperature at common cell temperatures which should result in substantial lithium creep deformation. Previous studies have characterized both the tensile creep properties of lithium foils[2] and compressive creep properties of bulk lithium rods.[3] Significant barrelling of the high aspect ratio lithium rods (initial height:width of 4:1 to 1:1) during compression made it difficult to determine the applied pressure on the lithium, obscuring the relationship between pressure and creep rate.[3] These important studies increased our collective understanding of the mechanical properties of lithium, but are not necessarily reflective of the compressive creep of lithium foils in a cell environment. Here, we report on the compressive creep of lithium foil within an electrochemical cell at commercially relevant stack pressures and temperatures.

Compressive creep deformation of low aspect ratio (1:40 to 1:10; initially 0.3 – 2.5 mm thick, 13 mm diameter) lithium metal foils was measured in hermetically sealed, rigid but flexible cells at temperatures between 30 - 110 °C and applied pressures between 1 - 2 MPa. Creep testing was performed by integrating a Conflat-style electrochemical cell[4] featuring welded bellows and an optical window with an Instron 5966 Universal Testing System (which measures / controls force and displacement). Creep rates were determined using the time-dependent displacement data after reaching a constant applied force. Two methods to convert force to pressure will be described, including the use of an inline camera to measure the lithium-compressive piston contact area.

Creep deformation was observed at all tested temperatures, and all pressures above 1 MPa. Creep rates were on the order of a few tenths of a micron per hour, which while small, implies very limited device lifetime with thin lithium layers. Pure lithium may not be physically strong enough for use in high energy density all solid-state devices, and alternatives (e.g. lithium-rich alloys) might need to be considered.

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