

Supplementary Methods:

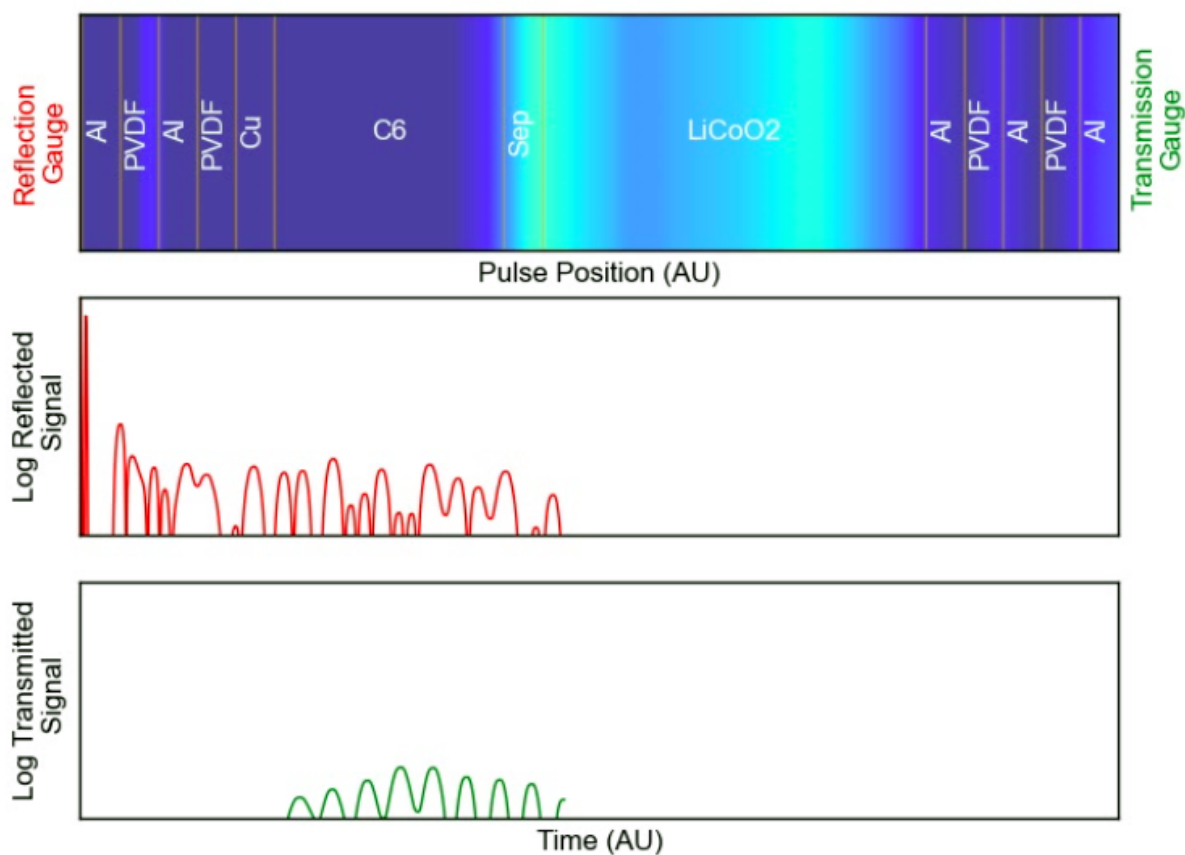
LiCoO₂ pouch cells were obtained in a pre-formed state from the Arnold lab at Princeton University. Cells had a nominal capacity of 160 mAh and a nominal voltage of 3.7 V.

Panasonic NCR18650B LiNiCoAlO₂ cells were obtained from a distributor, with a nominal capacity of 3400 mAh and a nominal voltage of 3.7 V.

Both Duracell and CVS brand Alkaline cells were purchased, with nominal capacities of 2850 mAh and nominal voltages of 1.5 V.

A Formlabs Form1+ 3D printer was used to design a cell holder for each battery geometry. The holders fit the batteries snugly, with cutouts for ultrasonic transducer placement. This allowed for easily repeatable experiments. Transducers were held in place using a 3D printed clip that applied light pressure and held the transducers perpendicular to the battery surface.

Supplementary Data:



Supplemental Movie | Clawpack simulation of an acoustic pulse traveling through a battery (single frame shown). The top plot shows a heat map of acoustic waves propagating through a LiCoO₂/graphite battery; the subsequent plots show the log of the reflected and transmitted signal intensities registered at the labelled extremes of the topmost plot. The movie shows the evolution of all three plots concurrently as the pulse propagates and echoes. This simulation is recalculated as a function of state of charge, where Dualfoil output files define both the geometry and density of the electrodes to create the results shown in Figure 2 and Figure S1.

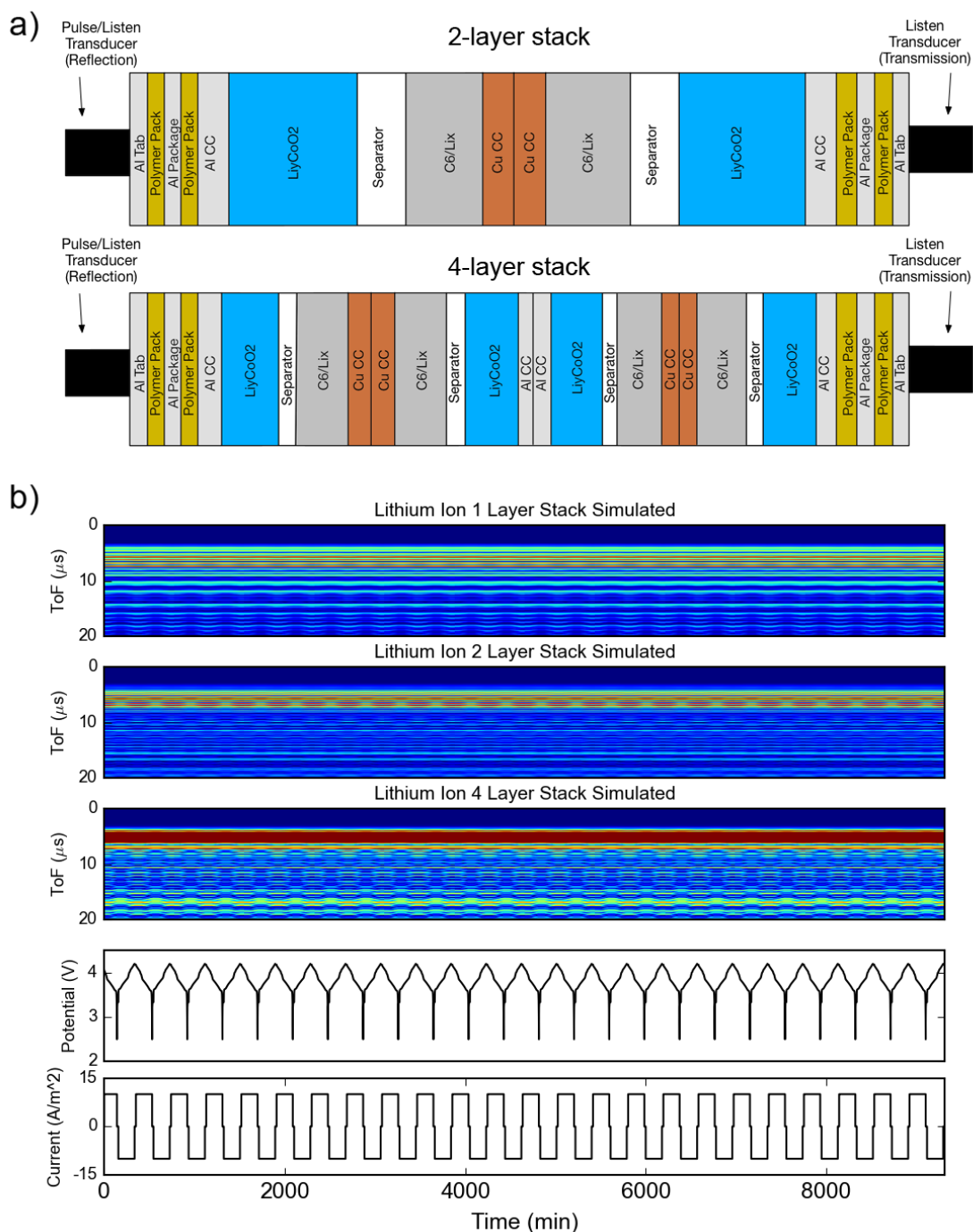


Fig. S1 | The effect of number of windings/layers on simulated transmission ToF. (a) schematics of the cell stacks used in the 2-layer and 4-layer Clawpack-Dualfoil simulations. **(b)** from top to bottom: simulated transmission ToF maps for 1-layer, 2-layer, and 4-layer stacks, simulated voltage profile, and simulated current profile. Of note is the increasing complexity in the transmitted signal with additional layers. The effects of interference and transmission/reflection at interfaces is increasingly pronounced in larger stacks.

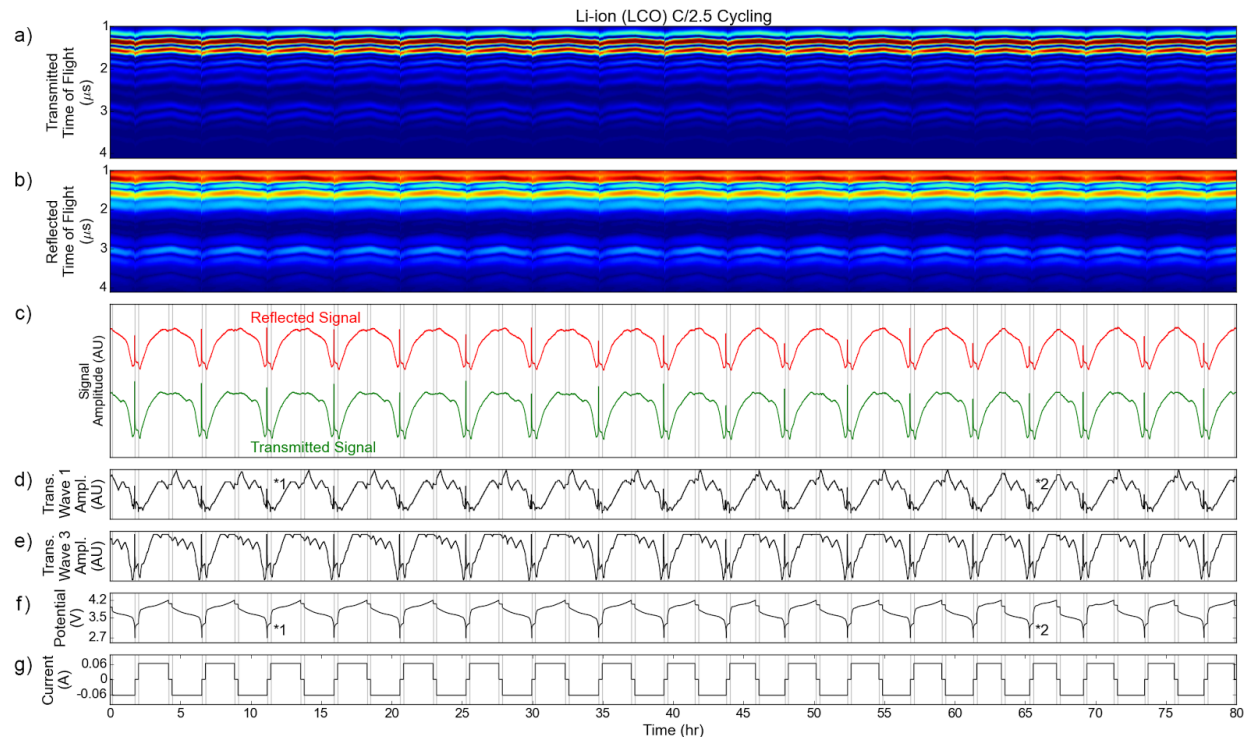


Fig. S2 | Full data set from Fig. 3: acoustic behavior of $\text{LiCoO}_2/\text{graphite}$ prismatic cell. (a,b) ToF maps for transmission and reflection modes, respectively, (c) total reflected (red) and transmitted (green) signal amplitudes, (d,e) traces for the amplitudes of transmitted waves 1 and 3, respectively, (f) cell potential, and (g) applied current as a function of cycling time. The vertical gray lines in panels c-g represent transitions between charge, discharge, and rest steps; arrows 1, 3, and 10 as well as markings *1 and *2 are discussed in the text.

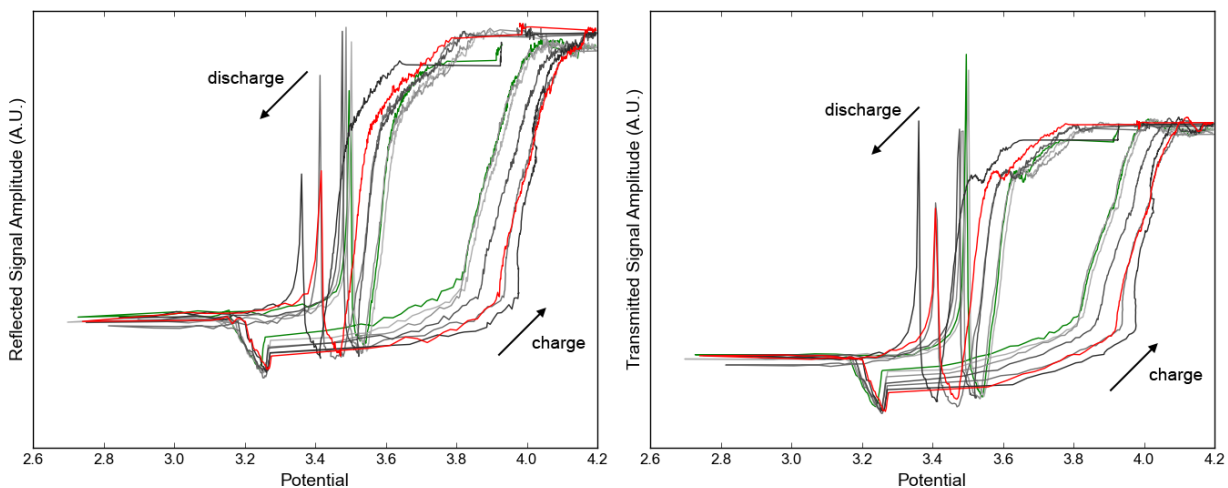


Fig. S3 | Acoustic signal amplitude as a function of cell potential. From the data down in Fig. S2, shown here are the (left) total reflected and (right) total transmitted signal amplitudes, plotted as functions of the cell potential during charge and discharge for every third cycle. The first cycle is indicated in green, with subsequent cycles shown as progressively darker shades of grey, and the final cycle indicated in red.

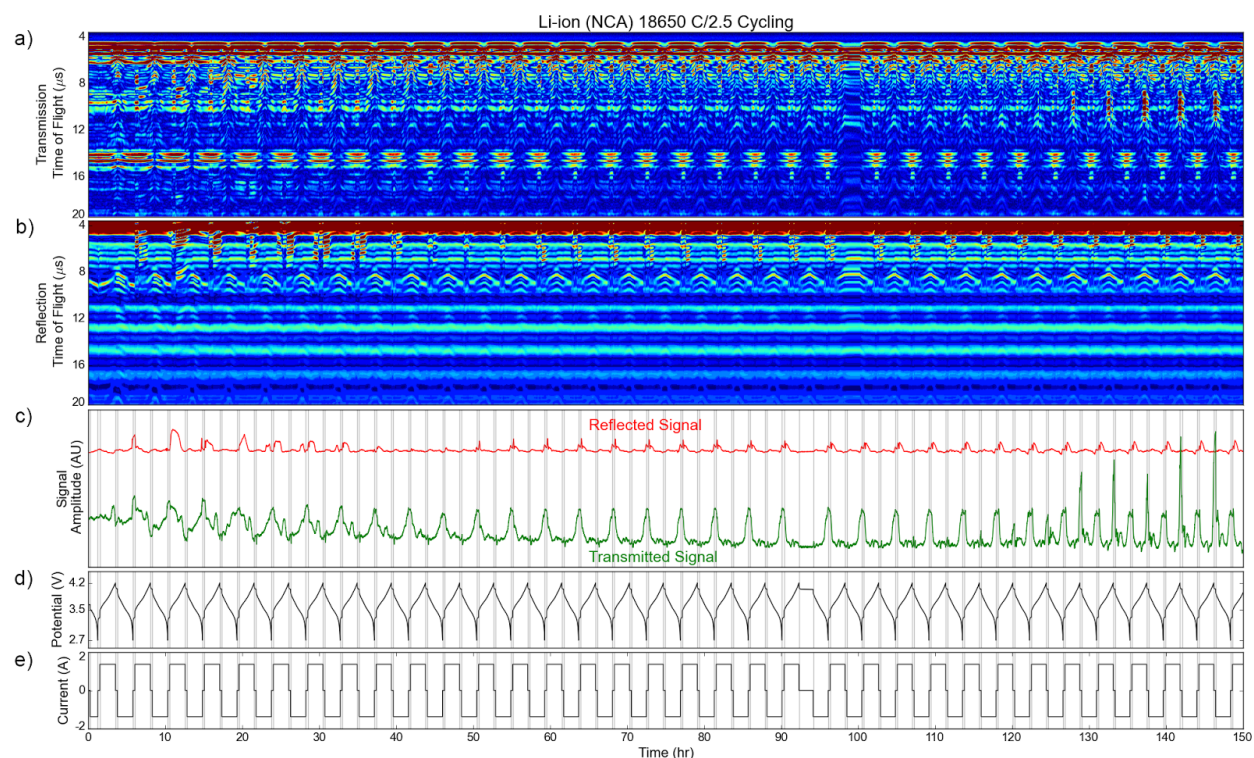


Fig. S4 | Full data set from Fig. 4: acoustic behavior of NCA/graphite 18650 cell. (a,b) ToF maps for transmission and reflection modes, respectively, (c) total reflected (red) and transmitted (green) signal amplitudes, (d) cell potential, and (e) applied current as a function of the cycling time. An extra one hour rest step was added around hour 96 of testing. The vertical gray lines in panels c-e represent transitions between charge, discharge, and rest steps.

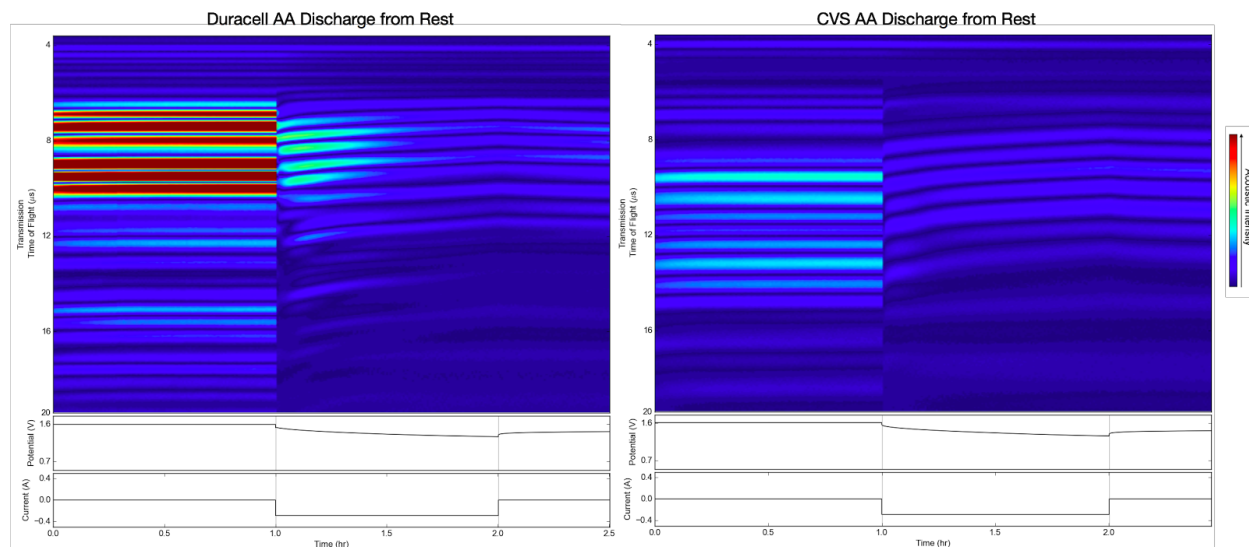


Fig. S5 | Acoustic comparison of Duracell and CVS pharmacy brand AA alkaline AA batteries.

These plots build on Figure 6 in the article; the change from equilibrium is shown for each brand. Note that the Duracell brand battery indicates a different ToF peak structure than the CVS brand cell. When taken apart, the Duracell has two major differences from the CVS brand battery: 1) the Duracell brand has a layer that we believe to be cellophane or similar between the anode and cellulose separator while the CVS brand does not; 2) the Duracell brand zinc has a metallic luster, whereas the CVS brand zinc anode is a duller gray. At the moment of discharge, the plots indicate a stark attenuation of the acoustic pulse in both cells, and the degree of attenuation appears to be stronger in the Duracell. We believe the attenuation in the Duracell is correlated with a loss of the corrosion inhibiting Duralock™ layer, as described in the main text.

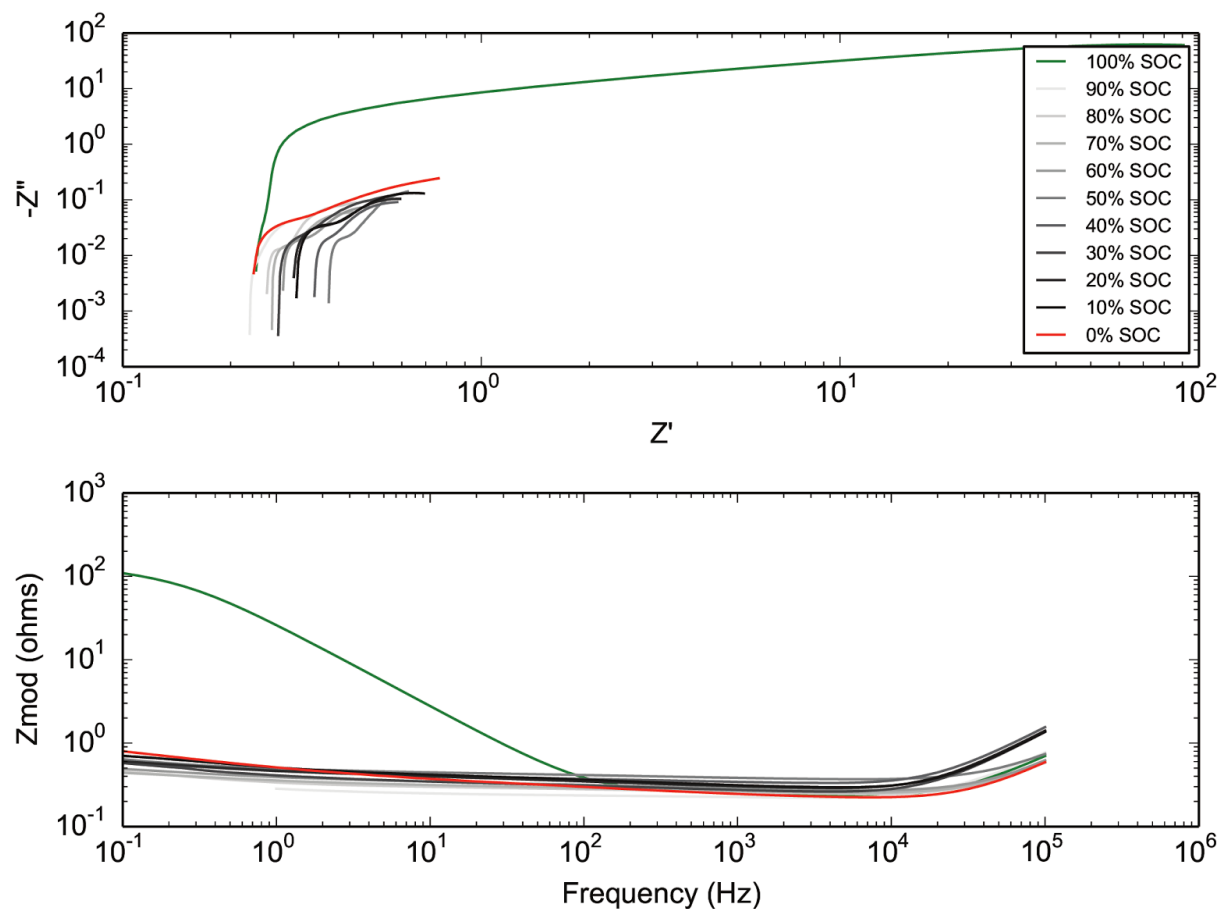


Fig. S6 | Electrochemical impedance spectra for a Duracell AA alkaline battery as a function of state of charge¹; Top: Nyquist plot. Bottom: Bode plot. The plots show the 2 order-of-magnitude drop in the total impedance in a AA alkaline battery. Subsequent EIS scans do not show appreciable changes based on state of charge. This correlates well with the sharp initial change in the transmitted acoustic signal shown in Figs. 6 and S8, and we posit that this is due to the breakdown of a corrosion inhibition layer that exists in Duracell batteries with proprietary Duralock™ technology.

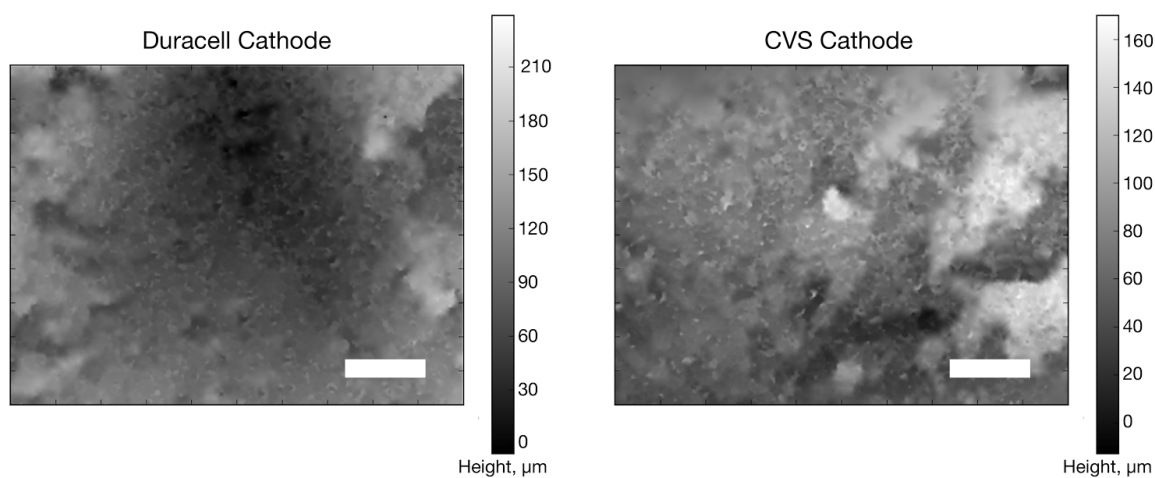


Fig. S7 | Confocal microscope images of Duracell and CVS brand Alkaline AA cathodes

Left: Duracell brand cathode. **Right:** CVS brand cathode. Scale bars are 250 μm for both. The images show the presence of larger agglomerates in the CVS cathode relative to the Duracell cathode. We hypothesize that the larger particles in the CVS cathode may increase scattering of the transmitted ultrasonic pulse as compared to the Duracell, which would result in greater attenuation of the ToF peaks observed between 4 and 8 μs in Fig. 6 in the main text.

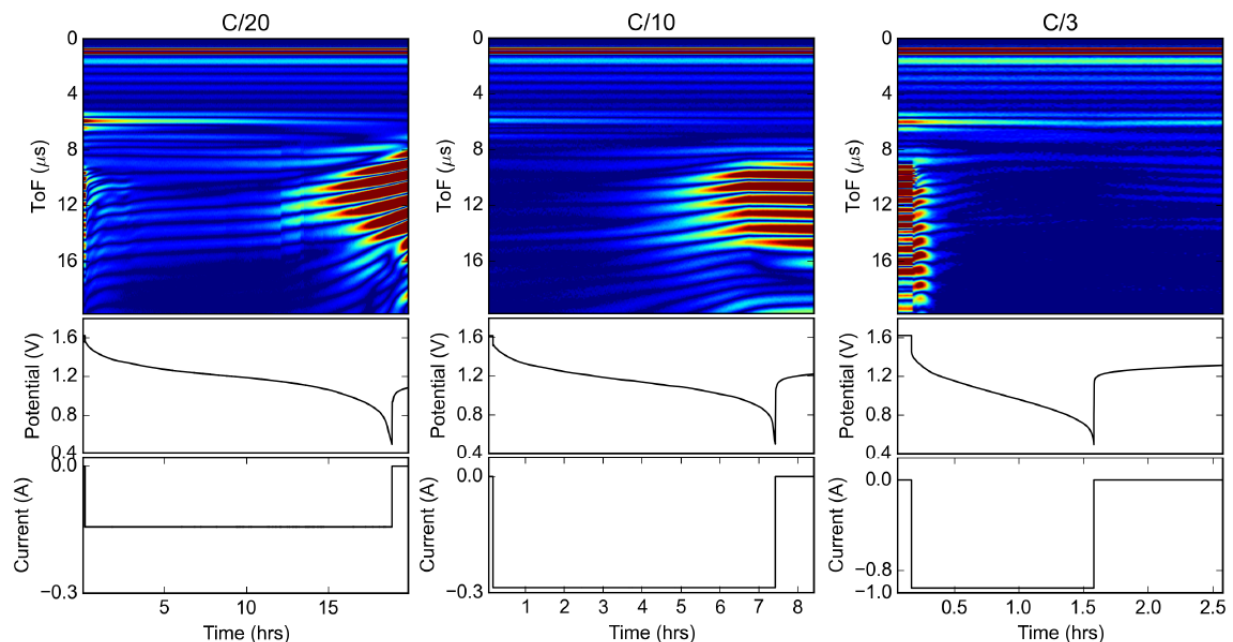


Fig. S8 | Acoustic evolution of a Duracell AA alkaline battery at multiple discharge rates; From left to right: C/20, C/10, C/3. The plots show the vastly different acoustic profiles at different discharge rates. At C/20 discharge rate, there are more high amplitude peaks evident at earlier time points, whereas at C/10, these peaks do not appear until much later in the discharge. The C/3 plot shows a very interesting phenomenon in which the cell appears to form a highly transmissive component immediately. This is posited to be a ZnO blocking layer at the separator/anode interface, which also results in premature failure.

Supplementary References:

1. S. Bhadra, B. J. Hertzberg, A. G. Hsieh, M. Croft, J. W. Gallaway, B. J. Van Tassell, M. Chamoun, C. Erdonmez, Z. Zhong, T. Sholklapper, and D. A. Steingart, *J. Mater. Chem. A*, 2015 (in review).