## In-Situ Raman of Few Layer Graphene Reveals Intercalation Staging for Potassium Ion Batteries Supporting Information





Figure S1. Differential capacity plot and corresponding charge-discharge curves of a potassium ion battery with a FLG electrode plotted vs SHE.



Figure S2. Picture of the *in-situ* Raman cell



Figure S3. a) 2D/G intensity scatterplot of carbonized nickel foam taken over a 40  $\mu$ m x 40  $\mu$ m area. b) G peak variation over the Raman map. The peak positions are normally distributed with an average position of 1582.4 cm<sup>-1</sup> and a standard deviation of 0.66 cm<sup>-1</sup>. The low standard deviation exemplifies the small spot to spot variation and justifies the use of a single point during the in-situ testing.



Figure S4. Galvanostatic rate study with 5 cycles at each of 50, 100, and 200 mA/g



Figure S5. a) Galvanostatic intermittent titration technique (GITT) using a galvanostatic pulse at C/10 (27.8 mA/g) for 30 minutes followed by a 1 hour relaxation (inset Figure S5).

The relaxation period in GITT allows the electrode to reach equilibrium, as seen by the potential plateau. This relaxation period can be used to assess the time it takes to reach equilibrium. For each relaxation period, the amount of time to reach 80% of the final relaxed voltage was calculated. This was plotted versus the final pulse voltage (Figure S5b). The maximum time in the voltage range used for Raman analysis is 300 sec, although the time to reach equilibrium during the main peak in the CV (<0.15 V) is below 10 seconds. During the 0.05 mV/s CV used for the in-situ Raman experiments, the maximum relaxation time of 300 sec (an overestimate) corresponds to a 0.015 V change, within the resolution of the spectra shown in Figure 3b. Below 0.075 V the relaxation time increases but by that voltage the metallic nature of the near stage I GIC interferes with the Raman spectra and doesn't provide additional information.



Figure S6. *In-situ* peak fit data for the 2D peak



Figure S7. Selective Raman spectra from the *in-situ* experiment with a larger range of wavenumbers. A D peak around 1336 cm<sup>-1</sup> is not observed during the *in-situ* study.