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## A Raman Spectroscopic Study of Graphene Cathodes in

### High-Performance Aluminum Ion Batteries

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#### ABSTRACT

Few layer graphene is a promising cathode material for aluminum-ion batteries that use chloroaluminate (AlCl<sub>4</sub>) ionic liquids as the electrolyte. A fundamental understanding of interactions between the few layer graphene cathode and the ionic liquid electrolyte is key for realizing the full potential of these systems. Through in situ Raman spectroscopy and density functional theory calculations, we show that the cathode is capable of achieving stage-one intercalation within the operating voltage window, leading to improved cell performance. We also show that the presence of structural defects in the few layer graphene such as pores induced via plasma exposure or nitrogen dopants can deteriorate the cell performance by either decreasing the electrical connectivity or precluding stage-one intercalation respectively. The cathodes made with highly crystalline few layer graphene display high power and energy densities (~200 Wh kg<sup>-1</sup> at 200 W kg<sup>-1</sup> and ~160 Wh kg<sup>-1</sup> at 5000 W kg<sup>-1</sup>), and are stable with no loss in performance up to 1000 cycles while fully charging to 2.4 V.

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