## Author's Accepted Manuscript

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 PII:
 S0926-2040(16)30004-2

 DOI:
 http://dx.doi.org/10.1016/j.ssnmr.2016.03.003

 Reference:
 YSNMR729

To appear in: Solid State Nuclear Magnetic Resonance

Received date: 15 January 2016 Revised date: 2 March 2016 Accepted date: 3 March 2016

Cite this article as: John M. Griffin, Alexander C. Forse and Clare P. Grey, Solid-state NMR studies of supercapacitors, *Solid State Nuclear Magneti Resonance*, http://dx.doi.org/10.1016/j.ssnmr.2016.03.003

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### **ACCEPTED MANUSCRIPT**

## **Solid-State NMR Studies of Supercapacitors**

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#### Abstract

Electrochemical double-layer capacitors, or 'supercapacitors' are attracting increasing attention as high-power energy storage devices for a wide range of technological applications. These devices store charge through electrostatic interactions between liquid electrolyte ions and the surfaces of porous carbon electrodes. However, many aspects of the fundamental mechanism of supercapacitance are still not well understood, and there is a lack of experimental techniques which are capable of studying working devices. Recently, solid-state NMR has emerged as a powerful tool for studying the local environments and behaviour of electrolyte ions in supercapacitor electrodes. In this Trends article, we review these recent developments and applications. We first discuss the basic principles underlying the mechanism of supercapacitance, as well as the key NMR observables that are relevant to the study of supercapacitor electrodes. We then review some practical aspects of the study of working devices using ex situ and in situ methodologies and explain the key advances that these techniques have allowed on the study of supercapacitor charging mechanisms. NMR experiments have revealed that the pores of the carbon electrodes contain a significant number of electrolyte ions in the absence of any charging potential. This has important implications for the molecular mechanisms of supercapacitance, as charge can be stored by different ion adsorption/desorption processes. Crucially, we show how in situ NMR experiments can be used to quantitatively study and characterise the charging mechanism, with the experiments providing the most detailed picture of charge storage to date, offering the opportunity to design enhanced devices. Finally, an outlook for future directions for solid-state NMR in supercapacitor research is offered.

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