

Sucre battery performance

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Rechargeable batteries that utilise lithium-ion or sodium-ion chemistry are important for applications including electric vehicles, portable electronics, and grid-scale energy storage systems1,2. While electrode design and the development of high capacity materials are relatively advanced, high-rate (power) performance still needs to be improved for a range of applications3. In particular, high rate performance is critical for rapid charging and high power delivery4.

Rate performance in batteries is limited because, above some threshold charge or discharge rate, RT, the maximum achievable capacity begins to fall off with increasing rate. This limits the amount of energy a battery can deliver at high power, or store when charged rapidly. Attempts to solve this problem have involved targeting the electrode5,6,7,8, the electrolyte9, and the separator10 with the aim of increasing RT and reducing the rate of capacity falloff above RT.

The factors effecting high-rate capacity are well known. For example, the rate performance can be improved by decreasing active particle size11,12,13, and electrode thickness14,15,16,17, or by increasing solid-state diffusivity11, conductor content7,16,18, or electrode porosity16,19, as well as by optimising electrolyte concentration14,16 and viscosity16.

Based on such information, it is accepted that rate performance is limited by: electronic transport in electrodes; ion transport both in bulk electrolyte and electrolyte-filled pores; solid-state diffusion of ions in the active materials and electrochemical reactions at the electrode/electrolyte interface12,20,21,22. One would expect that speeding up any of these processes would improve rate performance.

However, in practice, it is difficult to quantitatively link the observed rate performance to the factors given above. The most commonly reported experimental rate performance data are capacity versus rate curves. Ideally, the experimentalist would be able to fit his/her capacity-rate data to an analytic model which quantitatively includes the influence of the parameters above (i.e. electrode thickness, porosity, particle size, etc.). However, to the best of our knowledge, comprehensive, fittable, analytic models are not available.

To address these issues, we have developed a semi-empirical equation which accurately describes the rate dependence of electrode capacity in terms of electrode properties, via the characteristic time associated with

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charge/discharge. Importantly, we derive a simple expression for this characteristic time, which includes the mechanistic factors described above. Together, these equations accurately describe a wide range of data extracted from the literature.

This work was inspired by recent work on rate limitations in electrically limited supercapacitors32,33, which describes the dependence of specific capacitance, C/M, on scan rate, v:32

Here Q/M is the measured, rate-dependent specific capacity (i.e. normalised to electrode mass), QM is the low-rate specific capacity and t is the characteristic time associated with charge/discharge. Although we have written Eq. (2) in terms of specific capacity, it could also represent areal capacity, volumetric capacity, etc., so long as Q/M is replaced by the relevant measured parameter (e.g. Q/A or Q/V) while QM is replaced by the low-rate value of that parameter (e.g. QA or QV). Although this equation is semi-empirical, it has the right form to describe rate behaviour in batteries while the parameters, particularly t, are physically relevant.

To demonstrate that Eq. (2) has the appropriate properties, in Fig. 1 we use it to generate plots of Q/M versus R for different values of QM, t and n. In all cases, we observe the characteristic plateau at low rate followed by a power-law decay at high rate. These graphs also make clear the role of QM, t and n. QM reflects the low-rate, intrinsic behaviour and is a measure of the maximum achievable charge storage. Taylor-expanding the exponential in Eq. (2) (retaining the first three terms) gives the high-rate behaviour:

confirming a power-law decay with exponent n, a parameter which should depend on the rate-limiting mechanisms, with diffusion-limited electrodes displaying n = 1/2. Alternatively, by analogy with supercapacitors, other values of n may occur, e.g. n = 1 for resistance-limited behaviour32.

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