



# *In-situ* and *ex-situ* measurements of thermal conductivity of supercapacitors



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## ABSTRACT

Thermal signature of supercapacitors are investigated *in-situ* and *ex-situ* using commercial supercapacitors.

Regarding the *in-situ* method, four supercapacitors were connected in series, with thermocouples embedded between the supercapacitors. As the applied current was increased, the temperature measured at the intrinsic positions also increased. When cycling at a current density of  $0.11 \text{ A cm}^{-2}$  the centre temperature increased by 14 K compared to the stack surface temperature. This is an important figure as literature states that an increase of 10 K leads to a corresponding decrease in the lifetime by a factor of 2. Using the obtained temperature profiles, the effective thermal conductivity of the stack was found to vary between  $0.5 \text{ W K}^{-1} \text{ m}^{-1}$  and  $1.0 \text{ W K}^{-1} \text{ m}^{-1}$ , depending on the compaction of the stack.

For the *ex-situ* measurements, the thermal conductivity and the thicknesses of the supercapacitor material layers were measured individually in order to determine the corresponding thermal conductivity of the stack. When using this method an effective thermal conductivity of the stack of  $0.53 \pm 0.06 \text{ W K}^{-1} \text{ m}^{-1}$  was obtained. The analysis also demonstrated that the main contributor to the thermal resistivity and conductivity of the supercapacitor construction is the electrodes. This demonstrates that when managing heat from supercapacitors it is important to focus on the thermal conductivity of the components materials.

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## 1. Introduction

Energy management is a key factor for a prosperous human society. Supercapacitors, also known as electric double-layer capacitors or ultracapacitors [1], play a vital role in the technological evolution as their key attributes of high power density makes them preferred over batteries in a wide range of applications [2–4]. Today supercapacitors are found in devices ranging from regenerative braking systems in electric cars, aiding data storing in computers, as well as to accelerate wind turbines after a period with little wind, or to prevent electrical drop-outs in solar panels [5–8]. Moreover, combined DC (direct current) energy storage hybrids can supply better power quality and demand less space when containing supercapacitors [9] and micro grids with fuzzy powers can be stabilised [10,11].

A major advantage of supercapacitors is their high power density per mass compared to, e.g. batteries. A simple way to compare energy storage and power source devices is to compare them in a so-called Ragone plot; a log–log diagram with the energy density (in  $\text{k Wh kg}^{-1}$ ) plotted as a function of the power density (in  $\text{W kg}^{-1}$ ) [12,13]. The reason for the different performances of batteries and supercapacitors in the Ragone plot, lays in the nature of how they store the electric energy [14]. In batteries the energy is stored by using electrochemical reactions, whereas supercapacitors store energy purely by electrosorption of ions on the surface of the carbon electrodes [15]. This allows supercapacitors to obtain a high power density at the cost of a moderate energy density, whereas batteries display the opposite trend [16]. This fact implies that supercapacitors are well suited for applications where energy uptake and supply has to happen fast, i.e. power management, whereas batteries are better suited for long-term energy supply, i.e. energy management.

The emergence of supercapacitors is closely related to the development of carbon electrode materials, electrolytes, and the

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design of the supercapacitor units and systems [1]. Today commercial supercapacitors are based on organic electrolytes [17] and film electrodes made from a blend of porous carbon (usually activated carbon; 85–95 wt%), polymer binder (5–10 wt%), and additives to improve the electrical conductivity [18,19]. Several reviews on the electrode materials and electrolytes used in supercapacitors have been published over the last years [2,18,20–22], supplemented with best practice papers with guidelines for testing [23–25], studies on the degradation and ageing of supercapacitors [26,27], and perspectives on current trends and future potentials of this technology [3,19,28].

The *thermal* conductivity of supercapacitors has not been studied in such detail as the respective *electrical* conductivity. Some studies dealing with system cooling and thermal effects have been published [17,29–34], but only few deal with thermal gradients and thermal conductivity directly [31,33] applying values that are not obtained specifically for supercapacitor components. For the activated carbon component film electrodes, the dry thermal conductivity of graphite [35], graphene [36], and carbon nanotubes [37], among other carbon materials has been reported in the range of  $0.1\text{--}0.2\text{ W m}^{-1}\text{ K}^{-1}$ . This is the same range of values reported for activated carbon measured in air [38]. To our knowledge, only one study reports the thermal conductivity of various types of supercapacitor electrodes both dry and soaked in electrolytes [39].

In the light of scarce literature on the internal temperature handling of supercapacitors the aim of this study is to supply data on electrode heat management, and also to elucidate heat bottlenecks in design of supercapacitor systems. The paper will aid this process by studying the *thermal* conductivity, and temperature profiles, of commercial supercapacitor materials through *in-situ* and *ex-situ* methods.

## 2. Theory

In order to determine the thermal conductivity of the supercapacitors,  $\lambda$ , an *in-situ*, and an *ex-situ* experimental approach is applied in accordance with theory on transport phenomena described by Bird et al. [40]. The *in-situ* method, method I, seeks to determine the overall thermal conductivity of a stack of four supercapacitors connected in series using a sum of least squares method to fit the thermal conductivity to the internal temperature gradient, the ohmic resistance, and the geometry of the stack. The *ex-situ* method, method II, uses an apparatus and an experimental method developed by Burheim et al. [39,41–43] in order to determine the individual through-plane thermal conductivities of porous carbon particle based materials such like those in supercapacitor electrodes. The *ex-situ* method will serve as a reference for the *in-situ* method, as the thermal conductivity of the supercapacitor constituting materials can be ‘summed up’ to the overall conductivity of the capacitors. Combining these two techniques in a single study creates a unique framework for studying and evaluating the thermal conductivity from a sub-component level and up to a large scale level of several supercapacitor units.

### 2.1. Method I: in-situ measurements

This section is dedicated to define the thermal conductivity,  $\lambda$ , from a stack of supercapacitors used in an isothermal calorimetric study. The experimental equipment used for this method is displayed in Fig. 3, which will be more thoroughly described in the experimental section. The flux of internal energy from the stack of supercapacitors,  $\dot{U}$ , is defined according to the first law of thermodynamics, in Eq. (1):

$$\dot{U} = \nabla(\lambda \nabla T) + \dot{Q} = 0 \quad (1)$$

where  $\lambda$  is the thermal conductivity,  $\dot{Q}$ , is the volumetric ohmic heat generation, and  $\nabla$  is the cartesian vector differential operator. When a system is thermally insulated in the  $y$ - and  $z$ -directions, as in our experimental set-up, the heat fluxes can be considered to be a one-dimensional one and one is left only with gradients in the  $x$  direction, *i.e.*;

$$\lambda \frac{\partial^2 T}{\partial x^2} = -\dot{Q} \quad (2)$$

The volumetric heat of the supercapacitors can be averaged to Eq (3):

$$\dot{Q} = \frac{Q_{\text{total}}}{V} = \frac{RI^2}{wh\delta} \quad (3)$$

where  $Q_{\text{total}}$  is the total heat generated by the supercapacitors,  $V$  is the volume of the supercapacitor stack,  $R$  is the electrical resistivity of the stack,  $I$  is the current entering the stack from the cycling unit, and  $w$ ,  $h$ , and  $\delta$  represent the width, the height, and the thickness of the supercapacitor stack, respectively.

Fig. 1 shows a sketch of the four supercapacitors as they are used in the experiments. In Fig. 1 the numbering of the various temperature measurements are indicated, from  $T_1$  through  $T_5$ , with the maximum temperature measured at  $T_3$ . Of temperature profiles, displayed in the results, measurement spots are distributed symmetrically about the maximum temperature in the middle of the stack,  $T_3$ 's geometrical position is used as the origin of the model:  $x(T_3) = 0$ . The total thickness of the stack of supercapacitors is represented by the greek letter  $\delta$ . As illustrated in Fig. 1, the measurement points of the temperatures  $T_1$  through  $T_5$ , are set at equal

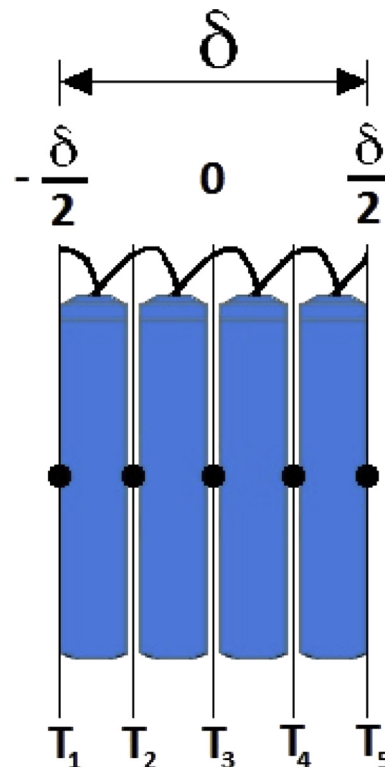


Fig. 1. Cross section of the capacitor stack with the placement of the thermocouples indicated.

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