Modeling Non-Ideal Behaviors of Supercapacitors' Equivalent Capacitance

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***Abstract*—Supercapacitors can improve energy storage performances and capabilities, in particular for applications requiring high specific capacitance and high specific power. Unfortunately, supercapacitors are characterized by several non-ideal and complex behaviors that must be modeled and taken into account. One of the most relevant issue consists in the definition of the equivalent capacitance and then to the energy that can be potentially stored and practically used. This paper discusses the possible capacitance definitions and measurement methods, including the possible approaches to model the capacitance and its non-idealities.**

***Keywords*—capacitance modeling, capacitive energy storage, energy management, energy storage, power supply, supercapacitor, ultracapacitor**

I. Introduction

Energy storage performances and capabilities can be improved and widened by a new class of devices known as supercapacitors (SCs), ultracapacitors, pseudocapacitors, double layer capacitors or electrochemical capacitors [1]. In particular, SCs are very suitable for applications requiring high specific capacitance and high specific power [2, 3]. Unfortunately, SCs are characterized by several non-ideal and complex behaviors that must be modeled and taken into account. Even though many papers can be found in literature about SC modeling [4-13], this continue to be a key topic with several open issue. One of the most relevant issue consists in the definition of the equivalent capacitance of a SC and then to the energy that can be potentially stored and practically used. This paper discusses the possible capacitance definitions and measurement methods, including the possible approaches to model the SC capacitance and its non-idealities. A specific focus was set on the voltage dependence of the SC capacitance, presenting several modeling and experimental results available in the literature.

II. SC Equivalent Capacitance

A. Non-Ideal Behaviors of SCs

Different studies on SC modeling can be found in literature. It is generally shown that a SC cannot be simply modeled by an ideal capacitance, not even taking into account parasitic resistances. The electric model must consider several important features shown by experimental data:

1. The SC behavior depends on voltage, frequency, current and temperature.
2. SC exhibits a complex dynamic over time which can be modeled in a general way using a several numbers of RC branches with a different time constant.
3. SC exhibits a self-discharge behavior.

B. General Definitions and Classifications

It is important to stress that the concept itself of capacitance is not trivial in a SC. In fact, different definitions and measurement methods can be adopted, as summarized in this section.

First, an important distinction between the differential, linear and integral capacitances must be introduced. The differential capacitance $c(v)$ is defined as the capacitance at a given cell voltage (the lowercase letters denote the variables quantities):

$$c(v) \triangleq \frac{dq}{dv} = i\frac{dt}{dv}. \tag{1}$$

The linear capacitance $C(v)$ can be defined as the capacitance of a linear capacitor storing the same charge Q of the SC at a given voltage [4, 6]:

$$C(v) \triangleq \frac{Q}{v} = \frac{1}{v}\int c(v)dv \tag{2}$$

The integral capacitance is defined by the stored charge Q over a voltage range ΔV:

$$C_{\text{int}} \triangleq \frac{Q}{\Delta V} = \frac{1}{\Delta V}\int_0^{\Delta V} c(v)dv = \langle c(v)\rangle_{\Delta V}. \tag{3}$$

The last equality shows that C_{int} corresponds to the mean of $c(v)$ over a certain voltage range. In practice, differential and integral capacitance are the same only if the voltage-time relation is linear and it is not the case of a SC.

The techniques for the capacitance measurement can be classified in 3 general categories [6]:

1. The methods based on AC impedance (on a frequency range) can be implemented by standard RLC meters and bridges, but specific instruments are commonly used to perform electrochemical impedance spectroscopy (EIS).
2. Cyclic voltammetry (CV) consists in a voltage sweep between two limit values while the current response is recorded, thus the area under these current curves is proportional to the charge stored.
3. Charge-discharge techniques: constant-current charging and discharging (CC CD) and constant power cycles are applied in order to characterize the cell voltage response. The IEC method and the Maxwell six steps method fall in this category of measurement as well as other charge-discharge methods where one or more current pulses are applied to the SC cell.

EIS techniques can measure only the differential capacitance while transient techniques (CV and charge-discharge process) can measure both differential and integral capacitance [9].

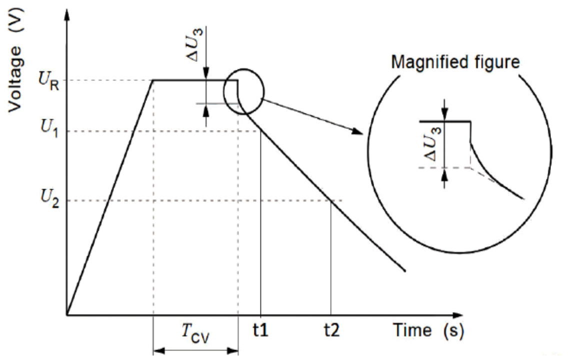


Fig. 1. Testing waveform according to Standard IEC 6239-1.

TABLE I. MEASUREMENT PARAMETERS ACCORDING TO THE DIFFERENT STANDARD METHODS

Procedure	Application	I_{cc}	V_1	V_2 (V)
IEC Class 1	Memory backup	C_r	$0.8V_r$	$0.4V_r$
IEC Class 2	Energy storage	$0.4V_rC_r$		
IEC Class 3	Power	$4V_rC_r$		
IEC Class 4	Instantaneous power	$40V_rC_r$		
Six-steps	DC	$I_5=0.1C_r$	V_4	$V_5=0.5V_r$

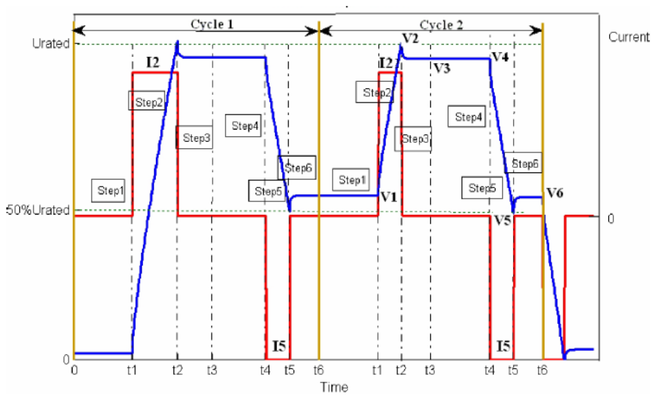


Fig. 2. Six-steps procedure adopted by Maxwell Technologies.

C. Standard Methods to Measure Capacitance

The IEC Standard 62391-1 [14] defines two methods to measure the capacitance of a SC. The most used method (method 1A) is summarized through the voltage waveform shown in Fig. 1. The SC is charged with a constant current until the voltage reaches the specified value, then the charging process continues with constant voltage. Afterwards, the SC is discharged with constant current I_{cc} defined in the standard based on the specific application as summarized in Table I. According to this method the measured capacitance is

$$C = \frac{t_2 - t_1}{V_1 - V_2} I_{cc}, \tag{4}$$

where the V_1, V_2, t_2 and t_2 symbols are defined in the standard as a fraction of the rated voltage V_r as described in Fig. 1 and Table I. Even if it is not specified, this is an integral capacitance.

The six-step procedure is adopted by Maxwell Technologies (the main SC manufacturer) to estimate capacitance, specifying that it is valid only in DC applications with an RC model [15]. Also in this case, the capacitance is identified by (3) using more cycles with the values specified in Fig. 2 and Table I.

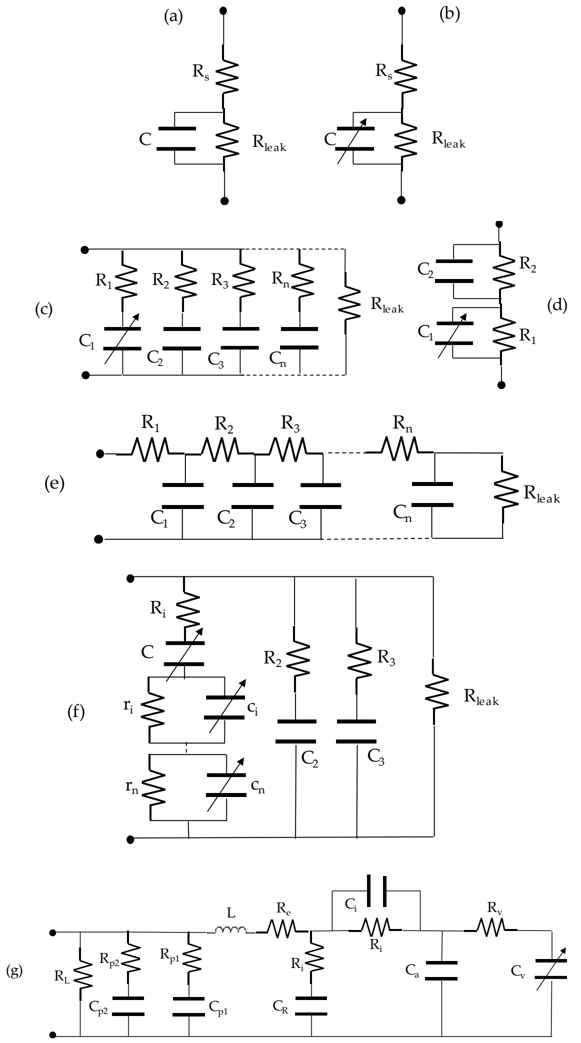


Fig. 3. SC equivalent circuits: simple classical circuit (a), classical circuit with voltage dependent capacitance (b), dynamic model with RC branches in parallel (c), simplified dynamic model (d), transmission-line model (e), Musolino model (f), Rafik model (g).

III. EQUIVALENT CIRCUITS FOR SC MODELING

Fig. 3 shows several SC models proposed in literature order by their complexity. The classical equivalent circuit in Fig. 3(a) and it consists of only 3 elements, with a series resistor representing the ohmic losses and a large parallel resistor modeling the self-discharge phenomena. This model can be built directly from manufacturer datasheets leading to a rough analysis of the real behavior. A more accurate model considers also the voltage dependence of the capacitance as shown in Fig. 3(b). The more complex models shown in Fig. 3(c)-(g) allow to capture the SC dynamic behavior on different time scales. These models are based on the transmission line theory associated with the distributed electric double layer [16]. The models in Fig. 3(f) [11] and Fig. 3(g) [8] were conceived to include all the possible SC non-idealities.

In any case, the first branch (R_1C_1) is used to model the immediate response while the other RC branches are set to have progressively higher time constants. Thus, the voltage variability is often associated to the first branch of the model and then to the immediate response of the SC.

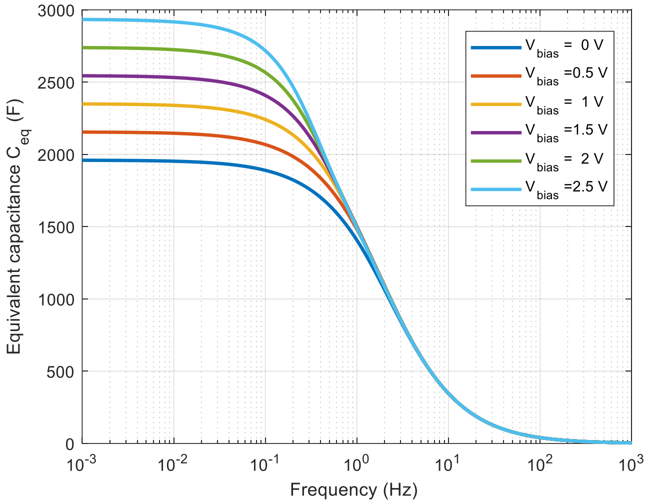


Fig. 4. Frequency response of the equivalent capacitance of a 2600 F cell at different voltage levels derived from the equivalent circuit in Fig. 3(g).

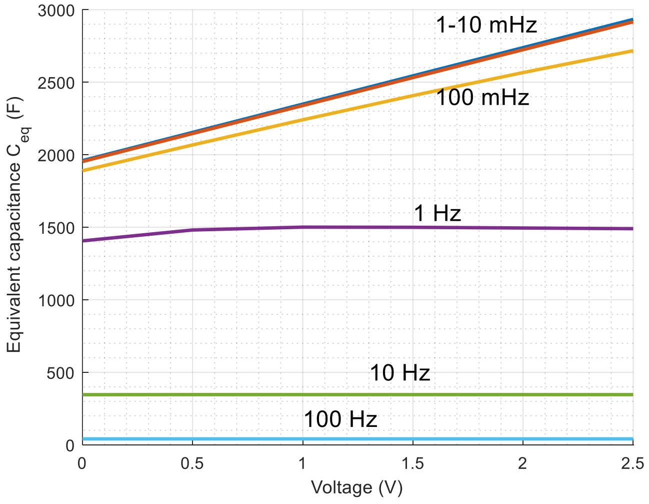


Fig. 5. Voltage capacitance dependence at different frequency derived from the equivalent circuit in Fig. 3(g).

The complexity and of the equivalent circuits lead many researchers to approach the problem by "fractional order models" based on non-integer differential equations and circuital elements [17]. This topic is too wide to be presented here. Nevertheless, it is useful to report that these models could effectively describe the SC dynamic behavior but without improving the modeling of the voltage dependence.

IV. Non-Ideal Behavior: Frequency Response

The frequency behavior of a SC must be considered in all the practical applications, as both equivalent capacitance and resistance of a SC depend on the operating frequency. The high-frequency (>1 kHz) behavior is important as SCs are coupled with power converters with switching devices. The frequency-dependent equivalent capacitance $C_{eq}(f)$ can be estimated from the SC complex impedance $Z(f)$ as:

$$C_{eq}(f) = -\frac{1}{2\pi \cdot f \cdot \mathrm{Im}[Z(f)]}. \tag{5}$$

Fig. 4 exemplifies the frequency and voltage behavior using real data derived from the model in Fig. 3(g). The dramatic capacitance fall around 1 Hz is due to the porous structure of the electrode and on the ion mobility [16].

As it will be described in next section, the $C_{eq}(f)$ values also depend on the SC voltage (charge). In this case, the measurements are normally performed by superimposing the small sinusoidal stimulus to a bias voltage V_{bias}. This is also exemplified in Fig. 4.

V. Non-Ideal Behavior: Voltage Dependence

A. Possible Definitions of the k_v Coefficient

The experimental data show that the SC capacitance depends on voltage [1]. This behavior descends from the electrochemical nature of SCs where an electric double layer occurs at the interface between a distributed and porous electrode and the electrolytic solution [8]. This nonlinear behavior is well visible by EIS measurements. As shown in Fig. 5, the capacitance values are strongly affected by V_{bias} at very low frequency (<1 Hz), while this phenomenon is less evident at higher frequencies where the effect of the diffusion into the inner region of the electrodes is no longer present. At these higher frequencies the equivalent capacitance is also lower, as shown in Fig. 4 and Fig. 5.

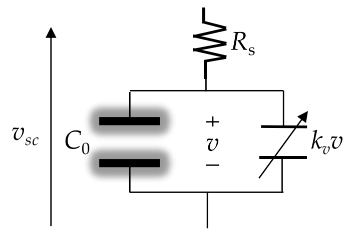


Fig. 6. Linear model of the capacitance of a SC by a standard capacitor in parallel to a variable capacitor proportional to the applied voltage.

Several authors proposed formulae to fit the experimental results for the voltage dependence of the SC capacitance [18]. The most noticeable analyses proposed hyperbolic [1, 19] or polynomial [20] approximations (where a_1, a_2, a_3 and V_x are fitting coefficients):

$$C(v) = \begin{cases} \left(\frac{1}{a_1} + \frac{1}{a_2 \cosh(a_3 v)}\right)^{-1} \\ a_1 + a_2 \tanh\left(\frac{v}{V_x} - V_x\right) \\ a_1 + a_2 v + a_3 v^2 \end{cases}. \tag{6}$$

Even though complex dependences as (6) were proposed, most of the researches [4] are based on a linear model like

$$C(v) = C_0 + k_v v \quad \text{with} \quad C_0 = C(v = 0), \tag{7}$$

where k_v, expressed in F/V, is a positive coefficient extracted from the experimental data. An equivalent circuit for this model can be as in Fig. 6. It is useful to stress that v in (7) should be the voltage across the capacitance and not that across the cell terminals (v_{sc}) that may include other elements.

The linear dependence can be expressed in other two forms depending on the voltage value used as reference:

$$C(v) = \begin{cases} C_{0.5} + k_v\left(v - \frac{V_r}{2}\right), & C_{0.5} = C\left(\frac{V_r}{2}\right) = C_0 + k_v \frac{V_r}{2} \\ C_1 + k_v(v - V_r), & C_1 = C(V_r) = C_0 + k_v V_r \end{cases} \tag{8}$$

In practice, any of the linearization (7) or (8) can be adopted, with the following properties:

1. The k_v value is the same.
2. The fixed capacitance (C_0, $C_{0.5}$, C_1 or others) must be reported to the reference voltage.

TABLE II. SUMMARY OF THE CHARACTERISTICS OF SEVERAL COMMERCIAL SC CELLS WITH A FOCUS ON THE APPROACHES ADOPTED TO INVESTIGATE AND MODEL THE VOLTAGE DEPENDENCE OF THE CAPACITANCE (REPORTED TO THE SUGGESTED PARAMETERS)

Cell	C_r (F)	V_r (V)	Measurement technique	$C(v)$ model	C_{given} (F)	C_0 (F)	$C_{0.5}$ (F)	k_v (F/V)	k_c (V^{-1})	k_{diff} (F/V)	Reference
Epcos 110F	110	2.7	CC CD	(6)	89	89	108.6	14.5	0.133	29	[7]
			CC CD	(6)	84.7	84.7	103.1	13.65	0.132	27.3	[7]
Maxwell BCAP140	140	2.7	EIS	(6)	99.1	99.1	117.4	14.7	0.125	29.4	[11]
Maxwell BCAP150	150	2.7	EIS	(6)	108	108	122.6	10.8	0.088	21.6	[11]
			CC CD	(6)	118.7	118.7	144.3	19.55	0.135	39.05	[10]
Epcos 200F	200	2.7	CC CD	(6)	158	158	195.9	28.1	0.143	56.2	[7]
			CC CD	(6)	152.7	152.7	192.3	29.35	0.152	58.71	[7]
Maxwell BCAP310	310	2.7	CC CD	(6)	282	282	344.1	46	0.133	92	[21]
Maxwell BCAP350	350	2.7	CC CD	(6)	237	237	298.4	45.5	0.152	91	[10]
			CC CD	(6)	232	232	293.2	44.5	0.153	89	[7]
			CC CD	(6)	234.7	234.7	290.3	41.1	0.141	82.2	[7]
	350	2.5	CV-EIS	(6)	241	241	294.1	42.5	0.144	85	[6]
Epcos 600F	600	2.7	CC CD	(6)	454.5	454.5	573.6	88.24	0.153	176.48	[7]
Maxwell BCAP1200	1200	2.7	CC CD	(6)	896	896	1062.6	70.47	0.056	140.95	[22]
			CC CD	(6)	981.6	981.6	991.1	60	0.071	120	[23]
			CC-CD	(6)	958	958	1093	100	0.091	200	[24]
Batscaps 1200F	1200	2.7	CC-CD	(6)	947	947	1068.5	90	0.084	180	[24]
Not reported	1500	2.5	Not reported	(6)	1125	1125	1312.5	149.55	0.114	300	[25]
Maxwell BCAP2600	2600	2.5	EIS	(6)	1960	1960	2203.8	195	0.088	390	[8]
Not reported	2600	2.5	Not reported	(6)	1975	1975	2300	249.95	0.113	500	[25]
Maxwell BCAP3000	3000	2.7	CC CD	(7)	2550	2408.3	2550	105	0.041	210	[12]
			CC CD	(7)	2768	2454.8	2768	232	0.083	464	[12]
			CC CD	(7)	2959	2613.7	3304.3	127.9	0.038	255.8	[26]
			CC CD	(6)	2366	2366	2672.5	227	0.084	454	[27]
SPSCAP 3000 F	3000	2.7	CC CD	(7)	2362	1996.5	2362	271	0.114	542	[12]
			CC CD	(7)	2407	2023.6	2407	284	0.117	568	[12]
			CC CD	(7)	2883.9	2245.6	3003.8	236.39	0.078	472.78	[26]
LSUC 3000 EA	3000	2.7	CC CD	(7)	2755	2056.2	2832.6	258.8	0.091	517.6	[26]
EATON XL60 3000F	3000	2.7	CC CD	(7)	2937	2633.9	3209.5	112.24	0.034	224.48	[26]
Not reported	3000	2.5	Not reported	(6)	1857	1857	2152.6	236.5	0.109	473	[28]
Skeleton SKELCAP SCA3200	3200	2.85	CC CD	(7)	2840	2673.3	2840	117	0.041	234	[12]
			CC CD	(7)	3133	2830.9	3133	212	0.067	424	[12]

B. *Practical Estimation and Modeling of k_v*

A variable capacitance has not the classical current-voltage relationship of a capacitor, but the general expression

$$i = \frac{dq}{dt} = \frac{d[C(v)v]}{dt}. \tag{9}$$

The derivatives in (9) depend on the specific behavior of the voltage dependence of the capacitance. In a circuital element linearized as in (7), the first derivative in (9) can be solved as

$$i = \frac{dq}{dt} = \frac{dv}{dt}(C_0 + 2k_v v). \tag{10}$$

This formula is also useful to model a SC with variable capacitance in software environments for the simulation of electrical circuits [24].

The different definitions of capacitance also lead to different expressions for its voltage dependence. The expression (7) strictly applies only for the linear $C(v)$ while for the differential capacitance the derivative in (2) yields:

$$c(v) = C_0 + k_{\mathrm{diff}} v \tag{11}$$

with

$$k_{\mathrm{diff}} = 2k_v. \tag{12}$$

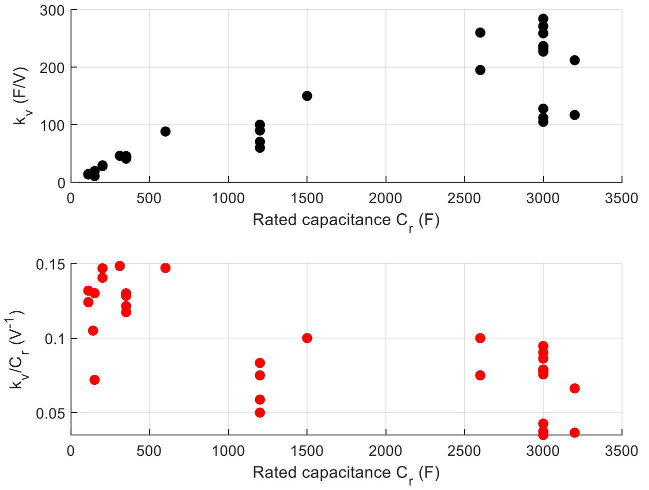


Fig. 7. Absolute and relative voltage coefficient k_v for the different SC cells listed in Table II.

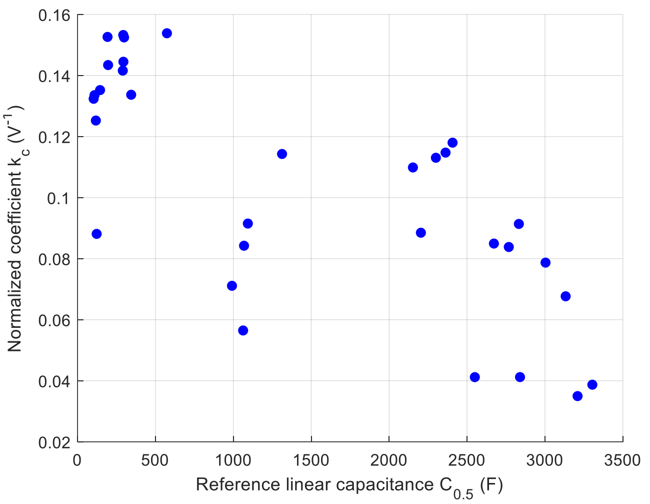


Fig. 8. Coefficient k_c for the different SC cells listed in Table II.

The adopted definitions lead to the following properties:

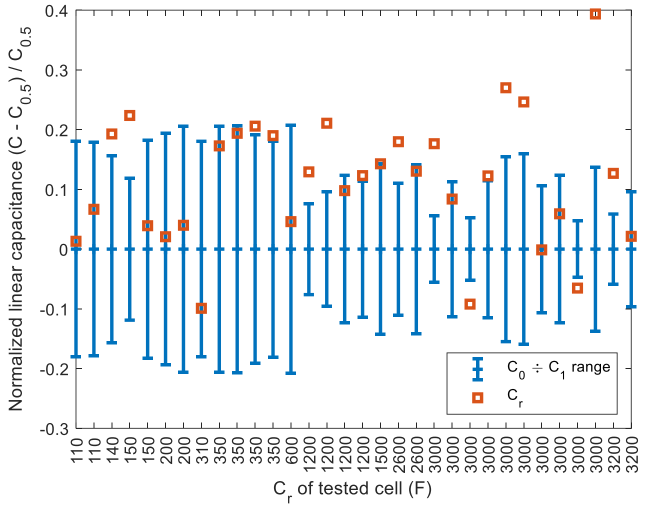
- The voltage coefficients k_{diff} and k_v are different.
- The capacitance at zero voltage is the same: $c_0 = C_0$.
- As it is always $c(v) > C(v)$ for $v > 0$, $c_{0.5} > C_{0.5}$ and $c_1 > C_1$.

For the CC CD techniques, the estimation of the voltage coefficients can be formalized by the time integration of (9) in order to obtain a time-voltage equation for a CC curve [7]:

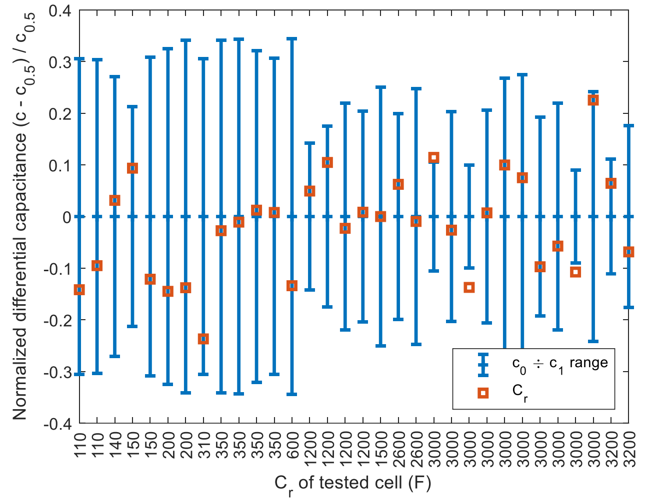
$$t = \frac{C_0}{I_{\text{cc}}} v + \frac{k_{\text{diff}}}{2 I_{\text{cc}}} v^2. \tag{13}$$

Since this equation contains 3 measurable quantities (t, I_{cc}, v) and 2 unknowns (C_0 and k_{diff}), the unknowns can be estimated by at least 2 measurement points [4, 7]. As stressed above, the voltage used in (13) must be that across the equivalent capacitance and not across the whole cell. In practice, this corresponds to subtracting the $R_s \cdot I_{\text{cc}}$ drop.

Since the measurements required by (13) may be characterized by significant noise, the use of a fitting procedure on more points can lead to more accurate estimations. In [12, 25], the estimation of C_0 and k_{diff} is carried out fitting several points measured on the charge-discharge curves over 4 consecutive CC CD cycles. Interestingly, these measurements discovered different values during the charge and the discharge processes, as reported in Table I.



(a)



(b)

Fig. 9. Comprehensive graphical tool to assess the voltage effect on capacitance, in case of linear (a) and differential (b) capacitance. The vertical bars delimited by C_0 and C_1 (c_1) provide a visual indication of the range spanned by k_c (k_c is higher for wider bars).

C. Survey of Researches on Voltage Dependence

Table II collects the results found in literature for the voltage dependence of the SC capacitance. In some cases, the same cell was tested by several authors with different techniques. The manufacturers' datasheets and the research papers may report a fixed capacitance C_{given} (that not necessarily coincides with C_r), usually without reporting the reference voltage. Since the data reported in Table II allowed the identification of the adopted conditions, they have been reported to the same voltage using (8) in order to obtain consistent comparisons.

The data in Table II were reorganized in Fig. 7, 8 and 9 for graphical analysis. In Fig. 7 the k_v and the k_v/C_r values are plotted against the C_r of the SC cells. The first impression is that k_v is proportional to C_r, but the observation of the relative values k_v/C_r rather suggests a decreasing with capacitance.

An even better trend can be observed in Fig. 8 where the use of $C_{0.5}$ instead of C_r introduces a new parameter k_c:

$$C(v) = C_{0.5}\left[1 + k_c\left(v - \frac{V_r}{2}\right)\right] \quad \text{with} \quad k_c = \frac{k_v}{C_{0.5}}. \tag{14}$$

D. *Graphical Control Tool*

Fig. 9 introduces a new comprehensive graphical tool. All the data are normalized to $C_{0.5}$ or $c_{0.5}$, so that $C_{0.5}$ or $c_{0.5}$ correspond to the zero ordinate. The vertical bars are delimited by C_0 and C_1 (c_1) in order to show the range spanned by k_c. The C_{given} and C_r values should be included within the error bars (the C_r above C_1 could be justified by different adopted definitions).

We suggest to report exactly $C_{0.5}$ or $c_{0.5}$, also because the SCs mostly operate in the range $V_r/2 \div V_r$ and the linear approximation seems to be valid only in this range [8]. Nevertheless, other choices could be sound provided that they are well justified and supported by the proper information on the reference voltage and voltage coefficient.

VI. Discussion and Conclusion

An effective use of the SC technology is possible only after evaluating and modeling their non-idealities. In particular, manufacturers typically neglect to specify the voltage dependence. Even the researches dealing with this problem rely on different definitions and procedures.

This paper showed that the k_c values seems to decrease with capacitance. This result is consistent with the models and the few data available for the modules and packs consisting of more SC cells that will be addressed in a future paper.

However, the observed trends are rather weak and blurred by the different results found even for the same C_r. In particular, some data reported in literature seems affected by conceptual mistakes in the solution of (13).

The first step for a rational use of SCs should consist in defining the proper parameters and measurement procedures. We recommend to specify in some way the adopted definition (linear, differential, integral) in any reported SC capacitance value. Moreover, we suggest the adoption of $C_{0.5}$ (or $c_{0.5}$) and k_c as reference value for specifications. Nevertheless, several issues are open for future researches.

The dependence of the C_0 and k_v estimations on the applied I_{cc} is not well investigated in literature. In [13] it is shown that different values of I_{cc} lead to different $C(v)$, but it is useful to remember that even the capacitance values depend on I_{cc}.

In general, each of the SC non-idealities could be regarded as a sort of efficiency assessing the capacitance usable in a specific application with respect to the rated capacitance that is potentially available in the SC:

$$C_{\text{available}} = \eta_C C_r. \tag{15}$$

This applies for the frequency dependence, for the dynamic response, for the effect of the power converters for SC charge/discharge, but also for the voltage dependence, even with combined efficiency effects. Of course, such effects have a counterpart in terms of stored and available energies. These very important aspects will be addressed by the authors in future works and papers.

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