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MEASURED ICE CRYSTAL CAPACITANCES: THE FAILURE OF THE ELECTROSTATIC ANALOGY

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1. INTRODUCTION

The inclusion of ice crystal mass and mass growth rates is important to the improvement of GCM and cloud resolved models in addition to the interpretation of remote sensing data from ice clouds. The concept of an ice crystal capacitance is often presented as a way of characterizing ice particles in clouds, both in terms of growth or sublimation. The mass growth equation for ice crystals shown below models crystal growth in

$$\frac{dm}{dt} = \rho\frac{dV}{dt} = \frac{4\pi C\sigma}{A+B} \quad (1)$$

terms of ice supersaturation, σ (related to vapor density), terms describing heat and vapor conductivity (A and B), and a crystal "capacitance", C. In principle, this model assumes an analogy between an ice crystal in a vapor field with that of a conductor in an electric field. While this capacitance model of crystal growth has been in the literature for decades (a discussion of which can be found in standard texts like Pruppacher and Klett, 1997), it has never been extensively tested against actually observed crystal mass growth rates, mainly due to a lack of reliable growth data.

The capacitance theory is currently limited to only the most simple shapes which can be described mathematically, e.g. symmetric plates and columns. This has traditionally involved the descriptions of plates as oblate spheroids, and columns as prolate spheroids or simple cylinders. More recently, supposedly refined calculational approaches have been developed for symmetric bullet rosettes and solid and hollow symmetric columns (Chiruta and Wang, 2002 and 2005), yielding capacitance values larger than the spheroid approach. However, the majority of ice crystals observed in situ (Korolev et al., 1999 and 2000) and in the laboratory (Bailey and Hallett, 2002 and 2004a,b) rarely assume simple shapes and are often quite irregular in appearance.

Experiments in recent years (Bailey and Hallett, 2002 and 2004a,b) have produced a substantial body of crystal growth results which have been shown to be in good agreement with in situ observations of crystal habit and dimensions (Bailey and Hallett, 2004b), in situ measurements of crystal mass remaining out of reach of current instrumental techniques. In these studies, volume growth rates, dV/dt, were determined which are related to mass growth rates via ice crystal density. When laboratory measured growth rates are inserted into the growth equation in order to determine crystal capacitances,

$$C = \rho\frac{dV}{dt}\frac{(A+B)}{4\pi\sigma} \quad (2)$$

capacitances significantly lower than those predicted by any of the current calculational approaches are obtained. The measured capacitances show that actual crystal growth rates are typically a factor of 3 to 10 times lower than those predicted by the capacitance model, indicating that the electrostatic analogy is not valid in terms of its description of crystal shape and its interaction with a vapor field. The electrostatic analogy fails for three main reasons.

1) The capacitance model assumes ice crystals have a symmetric shape (or distribution of symmetric components, as with bullet rosettes) and hence interact with a vapor field strictly according to the details of shape. The majority of even simple hexagonal ice crystals (i.e. plates, columns, and bullet rosettes) observed in situ and in the laboratory are asymmetric in shape, often with facets with irregular steps or layers.

2) The distorted shape of most simple hexagonal ice crystals reveals the presence of defects which alters their shape from an ideal hexagonal symmetry. These defects also likely change the vapor pressure over a faceted surface due to stress or strain. This likely affects the mobility and attachment of water molecules migrating over stressed surfaces, affecting what has been characterized as a "condensation coefficient". Hence, apparently equivalent facets on the same crystal are often observed to grow at different

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rates, resulting in scalene hexagonal plates and "flattened columns" (Bailey and Hallett, 2004), plates and columns which are not symmetrically hexagonal in cross section.

3) Laboratory observations of the growth of even simple plates and columns always reveal a spread of crystal diameters and thicknesses or lengths for crystals growing under the same conditions of temperature, ice supersaturation, and pressure (related to vapor diffusivity). This is also seen with in situ observations, though growth conditions in the atmosphere are less well defined and are more variable due to atmospheric motions. Again, this spread in growth characteristics is likely a reflection of the role of defects in determining the details of crystal growth, a factor which cannot be addressed by the capacitance model.

The measurement of mass growth rates in the laboratory provides a means of parameterizing crystal growth in terms of a more accurate "effective capacitance" which can be applied to the mass growth equation. This approach has the advantage of being applicable to more complex shapes which typically comprise approximately 95% or more of in situ (Korolev et al., 1999 and 2000) and laboratory grown crystals, the shapes for which are beyond current calculational approaches. Results for both simple and complex ice crystal shapes are presented and discussed.

2. EXPERIMENTAL DESCRIPTION

The growth data used to calculate capacitances in this work come from a crystal growth study (Bailey and Hallett, 2002 and 2004a) where a static diffusion chamber was used to grow ice crystals between -20 ^{o}C and -70 ^{o}C. Crystals were nucleated and grown on 30 um diameter glass filaments under controlled conditions of temperature, pressure, and ice supersaturation. Crystal growth was measured by analysis of time lapse video, and the glass filament substrate could be rotated so that accurate volume, area, and length measurements could be performed. A full description of the details of the method can be found in Bailey and Hallett (2002 and 2004a).

One criticism of static diffusion chamber results is the assumption that crystal habit may be affected by substrate interaction. Nucleation effects on habit is a factor with some laboratory methods, and a comprehensive assessment of these effects can be found in Bailey and Hallett (2002). However, fine glass substrate experiments consistently produce the habits observed in situ with only minor differences.

Another criticism applies to the situation when crystals are grown with high concentration, leading to vapor "shadowing" by crystals which grow close to each other on the filament, possibly reducing observed growth rates. At low ice supersaturation, it is possible to limit crystal concentration through control of initial nucleation. At higher ice supersaturations, nucleation can still be somewhat controlled, but crowding can become a problem. However, some crystals, which nucleate well before others, get a head start on growth, and often end up extending well beyond the filament substrate which can be crowded. In these cases, little or no shadowing occurs. All the growth data reported in Bailey and Hallett (2002 and 2004a) was obtained from crystals that essentially met this criteria, and only ice supersaturations which allowed good spatial separation between neighboring crystals was used for the analysis in this work.

3. CRYSTAL CAPACITANCES

The oblate and prolate spheroidal descriptions of the capacitance of simple plates and columns can be found in Pruppacher and Klett (1997) and Wang (2002) and come from the earlier work of McDonald (1963). This approach uses the semi-major (a) and semi-minor (b) dimensions of the relevant spheroids as described in the following expressions.

(plates) $$C_P = \frac{a\varepsilon}{\sin^{-1}\varepsilon}; \qquad \varepsilon = \sqrt{1-\frac{b^2}{a^2}} \qquad (3)$$

(columns) $$C_C = \frac{A}{\ln[(a+A)/b]}; \; A = \sqrt{a^2 - b^2} \qquad (4)$$

McDonald additionally described columnar growth in terms of a circular cylinder capacitance which can be found in Wang (2002). Chiruta and Wang (2002 and 2005) have more recently presented new capacitance descriptions which yield values larger than the spheroid calculations which are shown in figure 1. However, as will be shown, the oblate spheroid results already severely over predict the crystal capacitance, so larger values do not represent an improvement.

The capacitance values calculated from laboratory mass growth measurements, along with the theoretical values of McDonald, are presented in table 1 for plates and table 2 for columns. These values were taken from Wang (2002) where a dimensionless form of these parameters is presented. In the case of columns, circular

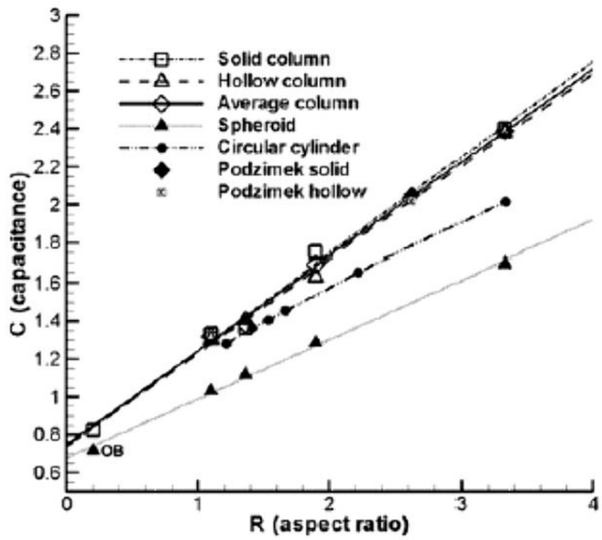

Figure 1. Ice crystal capacitances calculated by Chiruta and Wang and others (Chiruta and Wang, 2005). The lowest values represented by solid triangles are oblate and prolate spheroid results.

cylinder results are presented for comparison, though these values differ insignificantly from the prolate spheroid results. In order to put the theoretical and the measured values on the same basis, capacitances divided by plate diameter (C/D) or columnar length (C/L) are presented in comparison with aspect ratio, thickness divided by diameter (t/D) in the case of plates, and length divided by width (L/D) in the case of columns. For the laboratory measured values, an average aspect ratio value typically spanning ±10% of the stated value is accompanied by a range of measured capacitances. A few individual values are also presented in the case of extreme aspect ratios.

Again, it should be noted that crystals essentially free from vapor shadowing were used in these calculations, which is especially the case for those results with the largest plate aspect ratios (t/D) and the smallest column aspect ratios (L/D) which were obtained from growth at low ice supersaturation (1-5%) where shadowing effects are negligible. Maximum ice supersaturations for

Table 1. Theoretical and Measured Hexagonal Plate Capacitances

Oblate Spheroid Plate Capacitances					Measured Capacitances					
					-20 °C, 550 mb		-30 °C, 400 mb		-40 °C, 300 mb	
Diameter	thickness	t/D	Capacitance	C/D	t/D	C/D	t/D	C/D	t/D	C/D
2.0	0.225	0.112	0.730	0.365	0.84	0.27	0.83	0.11-.23	0.82	0.07-.29
2.0	0.177	0.089	0.698	0.349	0.63	0.15	0.73	0.36-.62	0.72	0.11-.18
2.0	0.126	0.063	0.664	0.332	0.20	0.10	0.61	0.16-.28	0.59	0.10-.36
2.0	0.103	0.052	0.648	0.324	0.13	0.02	0.40	0.07-.17	0.45	0.09-.26
2.0	0.086	0.043	0.637	0.319	0.12	0.08	0.32	0.09-.29	0.32	0.08-.15
2.0	0.072	0.036	0.628	0.314	0.08	0.07	0.20	0.02-.13	0.20	0.02-.13
2.0	0.064	0.032	0.622	0.311	0.06	0.03-.13	0.10	0.01-.12	0.08	0.04-.08
2.0	0.058	0.029	0.618	0.310	0.04	0.02-.04	0.06	0.02-.19	0.04	0.01-.11

Table 2. Theoretical and Measured Hexagonal Column Capacitances

Circular Cylinder Column Capacitances					Measured Capacitances					
					-30 °C, 400 mb		-40 °C, 300 mb		-50 °C, 250 mb	
Diameter	Length	L/D	Capacitance	C/L	L/D	C/L	L/D	C/L	L/D	C/L
2.0	2.85	1.42	1.36	0.48	1.00	0.30-.64	1.05	0.22-.57	1.04	0.27-.38
2.0	3.08	1.54	1.40	0.46	1.10	0.20-.37	1.10	0.29-.33	1.20	0.11-.22
2.0	3.33	1.66	1.45	0.44	1.25	0.17-.26	1.20	0.15-.55	1.40	0.09-.15
2.0	4.44	2.22	1.65	0.37	1.40	0.34-.36	1.70	0.08-.18	2.00	0.07-.11
2.0	6.67	3.34	2.02	0.30	1.60	0.09-.16	2.00	0.07-.15	3.00	0.05-.06
2.0	10.00	5.00	2.50	0.25	1.78	0.27	2.50	0.07-.09	6.50	0.02-.08
2.0	16.67	8.34	3.40	0.20			3.63	0.05	10.00	0.01-.05

plates were limited to 13% at -20 °C, 25% at -30 °C, and 40% at -40 °C, below water saturation in all cases. Maximum ice supersaturations for columns were limited to 13% at -30 °C, 25% at -40 °C, and 25% at -50 °C.

As can be seen from the tables, for the same or similar aspect ratios, the theoretical values over predict the capacitance by a factor of 3 to 8 in the case of plates (sometimes more), and approximately 2-4 in the case of columns. In both cases, deviation from the predicted value generally increases with decreasing aspect ratio in the case of plates and increasing aspect ratio in the case of columns.

Capacitances have also been calculated for bullet rosettes grown in the laboratory at temperatures below -40 °C. It has recently been established by both laboratory and in situ observations that bullet rosettes generally do not nucleate in the atmosphere for temperatures warmer than -40 °C to -36 °C, their appearance at warmer temperatures being due to sedimentation or fall from colder cloud regions aloft. These warmer rosettes undergo a considerable reduction in aspect ratio due to falling into a plate-like growth regime (Bailey and Hallett, 2004a and 2004b), additionally becoming hollow if fall occurs near water saturation or is enhanced to such values by ventilation. The laboratory examples used in these calculations are typical of bullet rosettes found in cirrus clouds at temperatures below -40 °C where individual bullets are generally solid. These values are compared in figure 2 along with theoretical calculations by Chiruta and Wang (2002) where the capacitance as a function of the number of bullets or lobes is compared. The calculations of Chiruta and Wang involve a mathematical formula that approximates hexagonal bullets as lobe-like structures. Here to it can be seen that the laboratory measured values are significantly lower than the theoretically derived values.

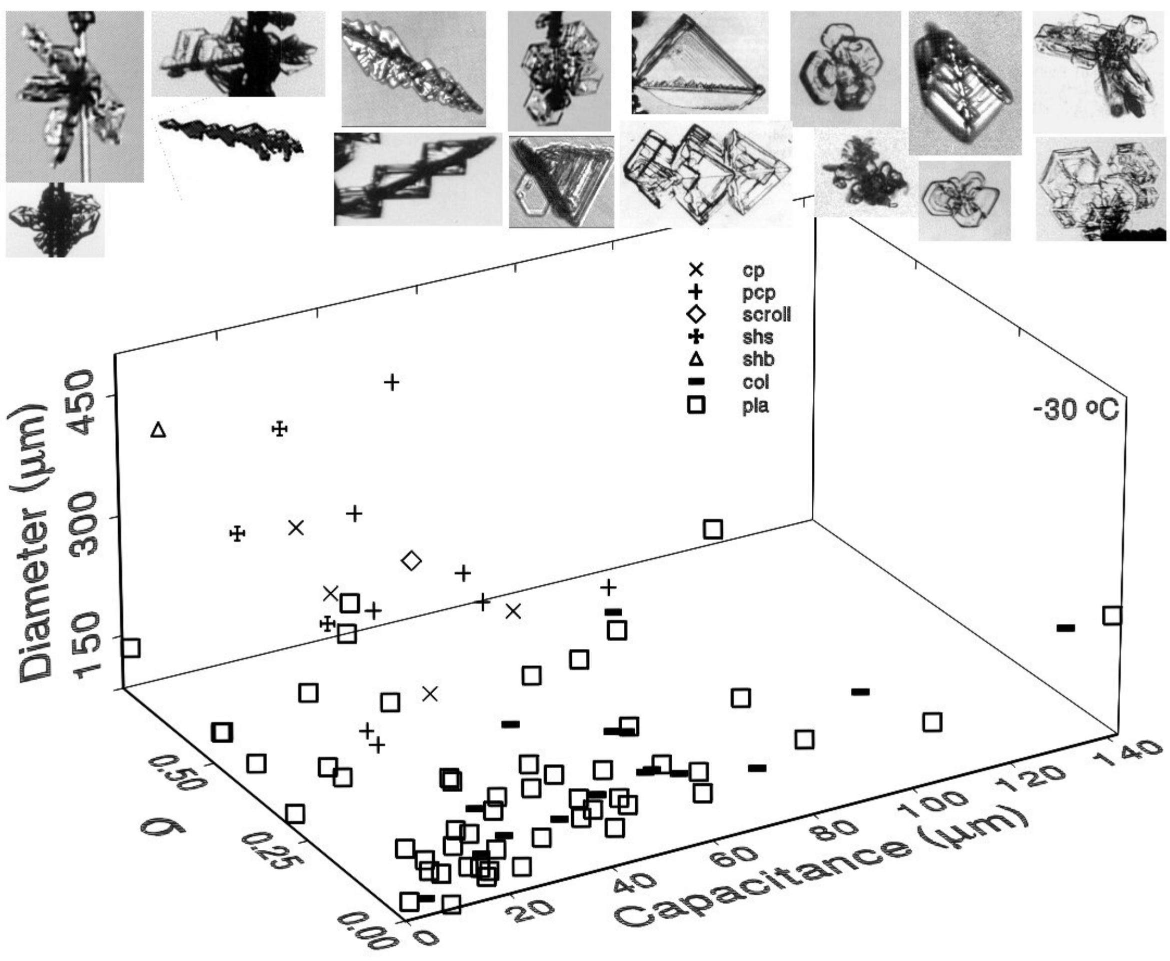

Figure 3. Laboratory measured ice crystal capacitances as a function of maximum dimension ("Diameter") for complex plate-like polycrystals like those shown at the top of the figure which include laboratory and in situ examples. None of the complex crystals shown are aggregates. A glossary of the of polycrystalline forms can be found in Bailey and Hallett, 2004a.

4. CAPACITANCE OF COMPLEX CRYSTALS

The utility of laboratory measured capacitances is especially evident when applied to complex shapes which are currently beyond any calculational approach. Below -20 ^{o}C the majority of supercooled cloud droplets freeze as polycrystals, an effect which increases with decreasing temperature. Hence below -20 ^{0}C, the majority of ice crystals are polycrystalline in form. Only at relatively low ice supersaturations are simple hexagonal plates and columns seen with any significant frequency. However even at low ice supersaturation, the majority ice crystals are compact polycrystals (Bailey and Hallett, 2002and 2004a,b).

In the static diffusion chamber experiments, the glass filament substrate could be rotated in order to view a crystal from any angle. Accurate bulk volumes and, hence, masses could be determined using a bulk ice density of 0.92 g cm^{-3}. Results for a sample of plate-like polycrystals typically observed at -30 ^{o}C are shown in figure 3 along with images of laboratory and in situ examples. It should be noted that none of the crystals in these images are aggregates. Plate-like polycrystals often exhibit pronounced complexity.

5. DISCUSSION OF RESULTS

One reason for the failure of the electrostatic analogy is the assumption of a high degree of symmetry in ice crystals. While this is expected from simple crystallographic considerations, the reality in nature is quite different. The vast majority of simple hexagonal plates and columns observed in situ and in the laboratory are not symmetric in cross section. Plates are typically scalene to some extent, having

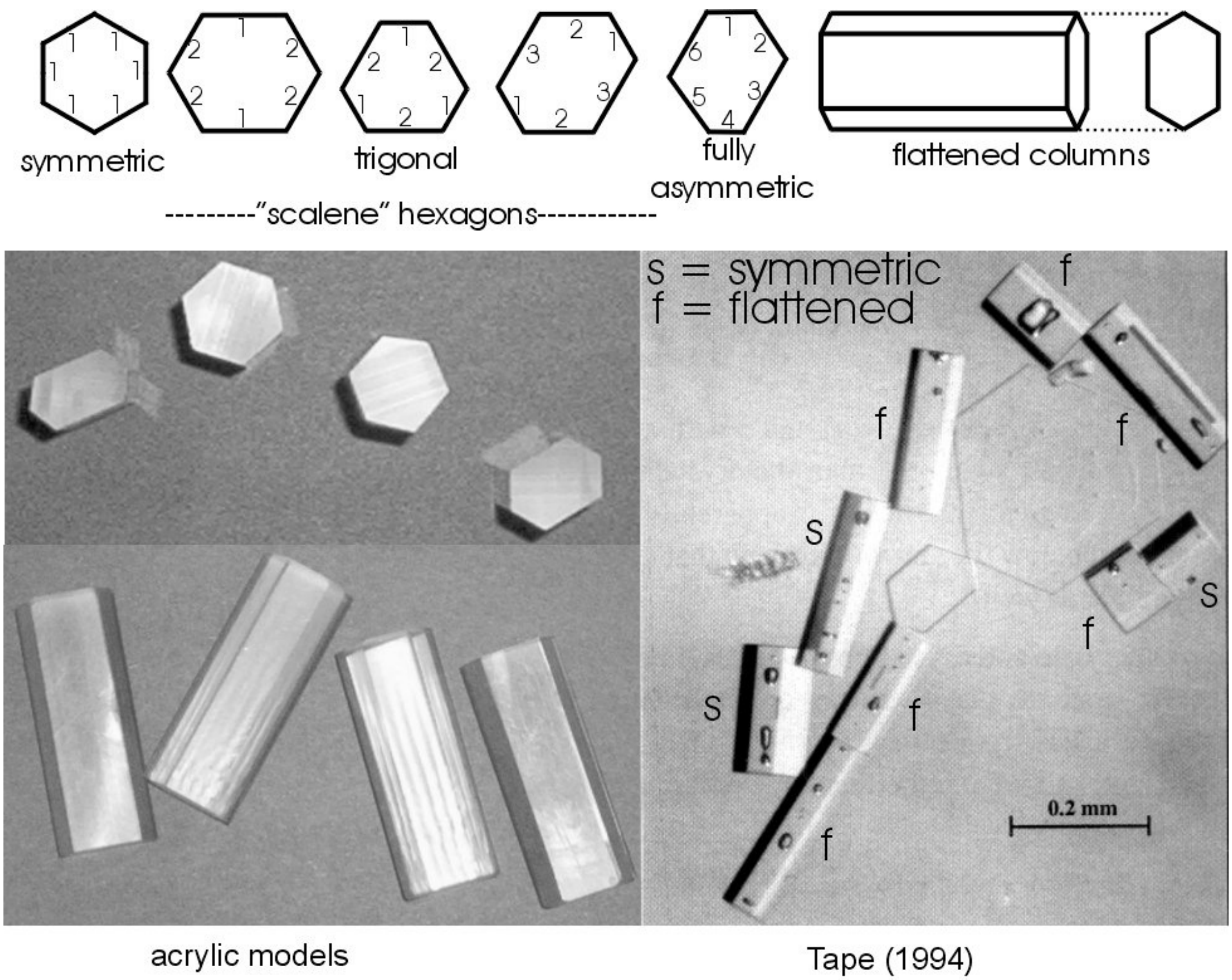

Figure 4. Asymmetric crystal shapes described as scalene plates or flattened columns. Numbers listed inside scalene plates label sides of equal length.

prism faces or sets of prism faces with different lengths which obviously grow at different rates. Flattened columns with similar cross sectional characteristics are depicted in figure 4.

The scalene character of plates has been easy to discern from laboratory and in situ data, but the scalene or flattened nature of columns has gone relatively unnoticed for some time. Scrutiny of data in the literature reveals it has always been there, but was probably mistaken for an optical illusion due to a particular perspective or viewing angle. However, considerations of how an asymmetric column appears in profile reveal that the flattened character is real. This is demonstrated in figure 3 where acrylic models of symmetric and asymmetric columns are shown end-on and in profile. For comparison, columns and plates gathered as "diamond dust" in the Antarctic by Tape (1994) are shown.

The pronounced asymmetry observed in most in situ and laboratory crystals below -20 ^{o}C is due to their predominantly polycrystalline nature which is determined by lattice defects and dislocations resulting in the formation of polycrystalline grain boundaries and related structures (Bailey and Hallett, 2004a). While it is more difficult to discern in simple plates and columns, defects and dislocations resulting in stressed or strained facets are likely the reason for the variability observed in these simpler forms, resulting in the prevalent scalene appearance of both.

6. CONCLUSION

The electrostatic analogy of the capacitance theory of ice crystal growth is highly flawed and does not produce the observed growth rates of ice crystals. It severely over predicts the growth rates in almost all cases involving even simple hexagonal shapes. The high degree of symmetry assumed in the calculations of crystal capacitances are not present in real in situ or laboratory grown ice crystals. Even if the capacitance model did correctly describe the growth of simple plates and columns, these habits form a small fraction of in situ and laboratory observed ice crystals which are dominated by polycrystalline forms, especially at temperatures below -20 ^{o}C, but also at warmer temperatures. The production and propagation of defects and dislocations in ice crystals are poorly understood but are likely the critical factors that determine habit details. These details are currently beyond any theoretical approach. However, laboratory measurements of mass growth rates can provide effective capacitances for even complex polycrystalline habits. In terms of growth parameters for model calculations, it is much more accurate to use effective capacitances measured in the laboratory which are substantially lower than those predicted by the capacitance theory.

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