Dielectric breakdown toughness from filament induced dielectric breakdown in borosilicate glass

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A R T I C L E   I N F O

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A B S T R A C T

The dielectric breakdown strength of borosilicate glass was measured as a function of the length of a conducting filament in order to determine the critical energy release for the growth of a breakdown channel. The concept is similar to the experimental determination of the toughness in fracture mechanics and based on a Griffith type model for the electrical energy release rate in dielectric materials with space charge limited conductivity. By Focused-Ion-Beam-milling and Pt-deposition, up to 100 μm long conductive channels were fabricated in 163 μm thick borosilicate glass substrates. The dielectric breakdown strength of substrates with filaments longer than 30 μm could be very well described by a 1/filament length dependence predicted by the model Schneider, 2013. With these results for the first time a critical energy release rate for dielectric breakdown was determined being 6.30 ± 0.95 mJ/m.

1. Introduction

Even though dielectric breakdown is a limiting factor for the reliability of electronic devices and components [2,3,4] and despite almost 100 years of research there is no commonly accepted understanding of this phenomenon. There is a great deal of effort being made to improve electrical performance in semiconductor electronics and batteries, but only a few research groups are concerned with the mechanism of dielectric breakdown itself. It seems to be clear that like for the mechanical strength and reliability it has to be distinguished between intrinsic and extrinsic failure mechanism [5]. Applying density function perturbation theory calculations (DFPT) it was shown that von Hippels avalanche model is able to predict the intrinsic breakdown for covalently bonded and ionic materials [6]. For the extrinsic breakdown the existing experimental results are more controversial. In this investigation we favour theoretical models based on the idea of a filamentary breakdown [1,5,7]. In such a model small conducting filaments of length z underneath the electrode in an electrical insulator of thickness d and a dielectric constant ε are assumed. These might be grain boundaries, chemical inhomogeneities with locally increased conductivities or injected charges. Under the application of the external homogeneous electric field E these tiny filaments lead to an electric field enhancement at the filament tip – similar to the mechanical stress field singularity in fracture mechanics, which triggers the dielectric breakdown (Fig. 1). In analogy to fracture mechanics it could be shown that this concept enables the formulation of an energy release rate \( G_{bd} \), which is the released electric field energy per filament extension [1].

\[
G_{bd} = \frac{\pi}{8} \varepsilon_0 a d E^3
\]  

(1)

\( \varepsilon_0 \) is the permittivity of free space. Dielectric breakdown occurs, when the energy release rate reaches a critical value which is \( G_c \).

\[
G_{bd} = G_c
\]  

(2)

Eq. (2) can be solved for the corresponding applied electric field at dielectric breakdown \( E_{bd} \):

\[
E_{bd} = \frac{1}{c} \sqrt[6]{\frac{6}{5\pi}} \sqrt{\frac{G_c}{\varepsilon_0 d}} \frac{1}{\sqrt{d}}
\]  

(3)

with \( c \approx \sqrt[3]{0.15} \). It has been shown in several publications that the extrinsic breakdown strength \( E_{bd} \) is proportional to \( 1/\sqrt{d} \) [5,8,9,10]. In addition, more recently it was shown that also the proportionality to \( 1/\sqrt{\varepsilon} \) holds for bulk ceramics and polymers [11]. These experimental results support the physical concept of this breakdown model.

Although the temperature dependent breakdown data from Hoshina et al. [12] up to 100 °C are best fitted by the Griffith-type energy release rate model when compared to an intrinsic and thermal breakdown model. A necessity for the result given in Eq. (1) is, that space charge limited conduction (SCLC) prevails in the electrically insulating material [11,13]. This could be shown for several ceramics and is a decisive difference to other breakdown models [11,14].

In this investigation we apply this theoretical concept to determine...
the dielectric breakdown energy release rate $G_c$ experimentally introducing well-defined artificial conducting channels of length $a$ and radius of curvature $r_0$ at the filament tip.

2. Material and methods

2.1. Material

To our knowledge there are no dielectric breakdown studies with artificially prepared conducting filaments similar to fracture toughness measurements with artificially introduced and sharpened notches [15, 16, 17]. Consequently the critical length of a filament as a function of the applied electric field is not known yet. The only hint we could use was our own publication [5] where we used the transition between extrinsic and intrinsic dielectric breakdown strength to estimate the size of the natural filaments of the order of some microns for ceramics. In order to avoid as much as possible natural defects such as cracks or grain boundaries as well as anisotropic material properties an amorphous glass was chosen as model material. The reason we did not use single crystals as in previous experiments [5, 18] is related to the fact that we observed preferential breakdown channel directions in the single crystals along certain crystal directions.

Commercially available borosilicate cover glasses were used. A summary of the physical characteristics can be found in Table 1.

Before every measurement the samples were cleaned with ethanol and dried at 60°C for half an hour. For conduction measurements until breakdown and breakdown tests, in addition, electrodes with conductive silver paint were applied, which is described in the following section in more detail.

2.2. Conductivity measurements

Electrical conductivity measurements were performed to determine whether the dielectric breakdown takes place in the space charge limited conduction (SCLC) regime. To cover a wide voltage range the conductivity measurements were divided in two parts. Conduction measurements up to 1 kV were performed in air with a more sensitive device and high voltage conduction measurements until dielectric breakdown were conducted in silicon oil to avoid partial discharge and flash over. All measurements were carried out at room temperature.

2.2.1. Conductivity measurements up to 1 kV

For these measurements a high-voltage meter Agilent 4339B was used. It can provide dc voltages up to 1 kV and detects currents from 60 fA to 500 pA. The measurement setup, consisting of three electrodes, is placed in a shielded resistivity cell (Agilent 16008B). With these three electrodes, namely a high-voltage (HV)-electrode with 18 mm and a ring-electrode with 22 mm diameter on same potential and the measuring-ele...
after the milling was finished the created channel was gauged. To determine the geometry of the milled channel, part of the sample had to be milled out additionally. Tilting the sample holder enabled to image the cross section of the filament. The filament’s geometry was gauged including the angle of inclination. This procedure was repeated for the different channel depths shown in Fig. 4, and thus a calibration equation was obtained. The obtained channels were always a factor 1.35 longer than the set milling depth. During the milling of the filaments, Pt had to be vapor-deposited on the entire visible surface from time to time. Due to the single beam, Ga⁺-ions, which are also used for imaging, slowly removed the originally deposited 5 nm thin gold layer, whenever an image was scanned. This Pt-deposition is only a few nanometers thick and has no effect on the milling of the filament. After the filaments were milled according to the parameters given in Table 2, the hollow channel was carefully filled with Pt to ensure a conductive filament without any cavities.

Table 2 summarizes the prepared channels displayed. The numbers in brackets are samples with additionally sharpened filaments. This means that after the filaments were milled, an extra fine tip was added to the first root tip applying the lowest ion beam current of 1 pA and the smallest point area with a radius of \( r_a = 50 \text{ nm} \) as shown in Fig. 3. These samples were prepared to check if the root tip is sharp enough as its radius of curvature has a huge impact on the electric field distribution at the tip.

Subsequent to the FIB-preparation for protection a droplet of silver paint was deposited above the filament. After drying for at least 30 min at 60 °C the gold layer was removed from the sample and the electrodes were applied out of conductive silver paint.

### 2.4 Dielectric breakdown test

For the breakdown tests the same high voltage source and electrode setup as for the conductivity measurements until breakdown was used but without the picoampere meter.

When measuring the FIB-prepared samples the conductive filaments were always placed on the negative or ground pole side, respectively, except from three samples with 50 μm filaments. These were placed reversed so that the filament faced the positive pole to examine whether electrons or holes are injected. The voltage was increased with a ramp of 0.2 kV/s until breakdown occurred. Breakdown was detected when the voltage signal suddenly dropped and a current flow arose. Experimentally, dielectric breakdown is detected when the Sefelec device detects a current flow and no steady voltage can be further build up any more. The current flow is limited to 2 μA due to an inner current...
limitation of the device. The dielectric breakdown event was always accompanied by noise emission and a breakdown channel. The dielectric breakdown voltage was defined as the maximum voltage right before the voltage collapse and the detection of a breakdown channel. As the voltage was detected with a rate of 250 ms the precision of the breakdown voltage is typically ± 50 V.

3. Results

3.1. Conductivity measurements

In Fig. 5, the measured current density $J$ is plotted as a function of the applied voltage $U$. The diagram includes the values of the low-voltage measurements from 1 to 1000 V and the high-voltage measurements from 1000 V until breakdown. The shown curve is the mean of four measurements on different glass samples. The slight step at 1 kV is due to the change of the equipment and is within the normal scatter of the data. Therefore, it can be assumed that even without ring electrode no or only very small surface currents were measured. In the lower voltage range, the measured data points describe a slope $s$ of approximately 1, which indicates ohmic conductivity in a double logarithmic plot. Beyond voltages of 2 kV, the rise of the current density increases and reaches a slope $s$ of $\geq 2$, indicating SCLC. The transition from ohmic to space charge limited conduction is quite sharp and the transition voltage $U_{tr}$ is around 3–5 kV. This behavior was already found for $\text{Al}_2\text{O}_3$ by Talbi et al. [14] and confirmed for $\text{TiO}_2$, $\text{BaTiO}_3$, $\text{ZrO}_2$ and $\text{AIN}$ by Neusel and Schneider [11].

The conduction measurements were performed with an average voltage ramp of 40 V/s which is 5 times slower than the ramp of 200 V/s for the breakdown experiments (see Fig. 6). The time to dielectric breakdown increases from approx. 240 s to 700 s. The dielectric breakdown at the end of the conduction measurements occurred on average at 28.0 ± 1.2 kV, which is significantly lower than the breakdown strength of 47.7 ± 4.7 kV during breakdown measurements. This time or ramp dependence of the dielectric breakdown strength is well known, but it was not possible to have the same ramp here as in the breakdown measurements since the conduction measurement needs time for current stabilization. As for the breakdown experiments with the 200 V/s ramp, a single breakdown channel can be seen after the conduction measurement until breakdown.

3.2. FIB-milled filaments

Because the sample must be destroyed to measure the geometry of the channel as described in Section 2.3, only channels used for the calibration can be seen here, not the actually conductive filaments, which were prepared for the breakdown measurements. Due to the standardized FIBing process, the images shown here can be considered to be representative for all samples. The FIB-milled filaments show a conical shape with a sharp pointed tip (Fig. 7). For all the calibration filaments with their different length varying between 10–80 μm the same ratio $\Psi$ between diameter $D$ and length $a$ of 0.15 was measured and a root tip radius $r$ of $\approx 500 \text{ nm}$. The filaments have a smooth surface and are homogeneously filled with platinum.

As known from fracture mechanics, the root tip radius has a big influence on the stress distribution and thereby on the fracture toughness. It is also known that there is a “critical notch root radius” for fracture tests and if this radius is exceeded, the calculated fracture toughness could be overestimated [20]. Also in electric fields, sharp edges are crucial. Therefore, a comparison of different root tip radii was made in addition. In Fig. 8 an example of a filament with a sharpened root tip is provided. The radius of curvature at this tiny extra tip is around 50 nm.

3.3. Dielectric breakdown tests

Fig. 9 shows the results of the breakdown tests of the sound (without artificial conducting filament) and the FIB-prepared glass substrates. The breakdown strength $E_{bd}$, defined as breakdown voltage per sample thickness, is plotted as a function of filament length $a$. All samples showed a single breakdown channel within the area of the electrodes. Samples where the breakdown occurred at the electrode edges were not included, because of the field enhancements at the electrode edges. As dashed line the prediction of the Griffith type energy release rate model is included. The sound breakdown strength is 292.87 ± 28.88 kV/mm, which is a scatter of about 10% standard deviation. Samples with 10 and 20 μm deep filaments were produced and tested as well, but these channels did not initiate breakdown, so the measured values were also counted as sound samples. In order to visualize these tests in Fig. 9 they are included with white rhombs. In samples with filaments equal to 30 μm length or longer, the breakdown was initiated at these FIB-
prepared conductive filaments. The samples with sharpened root tips showed no difference in comparison to the not sharpened ones as can be seen in Table 3. It is clearly seen that the breakdown strength decreases with filament length. In samples with 50 μm filaments but reversed polarity at breakdown test the dielectric breakdown was not initiated at the conductive filament. There was no influence of the filaments perceptible and the samples behaved like sound ones.

As the dielectric breakdown initiated at the artificially introduced filaments ≥ 30 μm, all breakdown strength results from filaments between 30–100 μm length were fitted by a least square fit assuming the $\frac{1}{a^2}$-dependence given in Eq. (3) and shown by the dotted grey line beyond $a = 30$ μm in Fig. 9. It can be seen that the experimentally measured dielectric breakdown strength fit well to the prediction. From the experimental results the calculated mean dielectric breakdown toughness $G_{bd}$ applying Eqs. (1) and (2) is $6.30 \pm 0.95$ mJ/m using the geometrical and physical values given in Table 1.

4. Discussion

On the working hypothesis that dielectric breakdown is initiated by tiny electrically conducting filaments dielectric breakdown measurements with artificially introduced conducting filaments were conducted

![Fig. 7. FIB images of the cross section of a FIB-milled filament used for calibration. a) Image showing the whole filament, completely filled with platinum. b) Magnification of the filament tip showing a tip radius of approx. 0.5 μm.](image)

![Fig. 8. FIB picture of a 10 μm filament with sharpened root tip. As this was a test filament, it was just roughly filled with Pt, which is seen at the bright region in the middle of the filament. Because the sharpened root tip is so small, it is difficult to picture it under a single beam FIB. While trying to focus the tiny tip under the ion beam the surrounding material got charged and additionally material was slowly removed and thereby the sharpened tip disappeared after a while. Therefore, we have no high quality image of the sharpened tip.](image)

![Fig. 9. Double logarithmic plot of the measured dielectric breakdown strength $E$ versus filament length $a$.](image)
Table 3
Mean dielectric breakdown strength and standard deviation for all different sample sizes. For 10 μm and 30 μm it is also distinguished between a 500 nm and 50 nm root tip radius.

<table>
<thead>
<tr>
<th>Filament length a [μm]</th>
<th>Root tip radius r [nm]</th>
<th>Breakdown strength E_{bd} [kV/mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>500</td>
<td>275.0 ± 16.5</td>
</tr>
<tr>
<td>50</td>
<td>295.5 ± 9.0</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>500</td>
<td>286.7 ± 0.0</td>
</tr>
<tr>
<td>30</td>
<td>500</td>
<td>270.9 ± 13.2</td>
</tr>
<tr>
<td>50</td>
<td>266.9 ± 6.0</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>500</td>
<td>248.9 ± 5.2</td>
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<tr>
<td>50</td>
<td>245.9 ± 14.4</td>
<td></td>
</tr>
<tr>
<td>60</td>
<td>500</td>
<td>230.3 ± 10.0</td>
</tr>
<tr>
<td>70</td>
<td>500</td>
<td>206.1 ± 8.6</td>
</tr>
<tr>
<td>80</td>
<td>500</td>
<td>186.0 ± 4.2</td>
</tr>
<tr>
<td>100</td>
<td>500</td>
<td>174.7 ± 8.5</td>
</tr>
</tbody>
</table>

in 163 ± 7 μm thick borosilicate glass plates. The results showed that filament lengths of up to 30 μm did not change the dielectric breakdown strength of 292.87 ± 28.88 kV/mm of the sound glass. Filaments with lengths of 10 and 20 μm were not sufficient for the initiation of breakdown and the experimentally observed breakdown channels were detected at different locations underneath the negative electrode. Thus, it can be concluded that there is a kind of “natural” filament length of up to 20 μm existing in the sound glass. Only for filament lengths of 30 μm and longer the breakdown could be detected at initiate at the filament. Hence only these filament lengths where fitted by a dependence and showed very good agreement, as predicted by Eq. (3).

From the known filament length and the obtained results for the breakdown strength, the dielectric breakdown toughness G_{bdw} was calculated as 6.30 ± 0.95 mJ/m². We point out that Eq. (3) was developed for a spherical specimen with a conducting filament in its center. Therefore Eq. (3) is only an approximation for small conducting surface lengths of 10 and 20 μm and a channel radius of 0.3-0.7 mm. The Grieth et al. [21] determined a critical length for di-electrical breakdown and the experimentally observed breakdown channels were detected at di-electrical breakdown as shown in Fig. 5. At low voltages the slope of the Griffith solution for cracks is only an approximation for surface cracks. Nevertheless the experimental results reveal dependence up to 100 μm filament length and we used these data for the evaluation of the breakdown toughness. Further electrical breakdown analysis is necessary to validate this approach.

This determined breakdown toughness of 6.30 mJ/m² is at the upper limit of estimations for Al₂O₃, TiO₂ and BaTiO₃ with 1.25 mJ/m², 2.21 mJ/m² and 4.22 mJ/m², respectively, based on the transition thickness between extrinsic and intrinsic dielectric breakdown [5]. Lin et al. [21] determined a critical J-integral between 2–40 mJ/m² for PZT (PZT807) in cylindrical specimen with 5–7 mm long initial channels and a channel radius of 0.3-0.7 mm. The J-integral evaluation is based on a perfect insulating dielectric material.

The Griffith type energy release rate model [1] assumes SCLC, which is found to be the dominant conduction mechanism in borosilicate glass before dielectric breakdown as shown in Fig. 5. At low voltages the slope of s = 1 of the data points indicates ohmic conduction in a double logarithmic J-U-diagram. Above a certain transition voltage U_{th} SCLC is the dominating conduction mechanism, meaning that more charge carriers are injected into the sample through the electrode than thermal carriers are present. This phenomenon is characterized by a slope of ≥ 2 in the double logarithmic plot of Fig. 5. For a perfect insulator with shallow traps on one energy level the current density of SCLC J_{SCLC} is exactly [8]

\[ J_{SCLC} = \frac{\Theta}{8} \frac{\Theta}{\Theta} \frac{\Theta}{\Theta} \frac{U^2}{d^2} \]

with the ratio of the free to the trapped carriers Θ (Θ = 1 for an ideal insulator without traps) and the charge carrier mobility μ. The distribution of the energy levels of the traps, J_{SCLC}=U², where s can be more than 2. From the experimental date the transition from ohmic to SCLC conduction occurs between 3 and 5 kV/m in this borosilicate glass.

Thus, the conductivity measurements show a similar behavior as those on Al₂O₃ by Talbi et al. and Neusel et al. [14,11]. In these experiments it could be shown that Schottky and Poole-Frenkel conduction can be excluded as dominating conduction mechanism. The fact that breakdown tests with reversed polarity don’t show breakdown initiation from the positively charged artificially introduced filament is an indication that electron injection is the source of the dielectric breakdown in the used borosilicate glass. These and previous findings support the notion that models, which are based on ohmic conduction, are not appropriate to describe the dielectric breakdown correctly.

In order to investigate whether the radius of curvature of the filament tip influences the dielectric breakdown strength for filaments with 10 and 30 μm length tip radii of 50 nm were FIBed. For the 10 μm long filaments, a sharper tip radius of 50 nm did not cause the breakdown to be initiated at the filament tip. For the 30 μm long filament, breakdown initiated at the filament tip but the dielectric breakdown strength did not change significantly in comparison to the 50 nm sized tip radii. As sharp edges and points tips are well known to increase the electric field strength, this result is astonishing. In a perfect dielectric the electric field E at the surface of a charged sphere of radius r scales with E ∝ 1/r and a field enhancement of a factor of 100 is expected decreasing the filament tip radius from 500 to 50 nm. If SCLC prevails, the electric field at the surface of a conducting sphere is proportional to E ∝ 1/r² [13], meaning that a 10 times smaller filament tip radius leads to a roughly 3 times higher field. The comparison of the 2 scenarios is in favour of a space charge mechanism in the sense that the local field strength at the filament tip is not decisive for breakdown but the electric energy released during filament extension.

5. Conclusion

The dielectric breakdown toughness for borosilicate glass was determined for the first time. The evaluation is based on SCLC as the dominant conduction mechanism right before breakdown. It was shown that FIB-milled filaments with lengths up to 20 μm do not initiate breakdown. Hence it is concluded that “natural” filaments of this size should exist underneath the electrodes. By changing the filament tip radius we could demonstrate that tip radii of 500 nm are small enough for a tip radii independent result. The FIB-milled channels are narrow and sharp enough to serve as model filaments and the extrinsic dielectric breakdown strength follows the 1/r²-dependence as predicted from a Griffith-type breakdown model. We assume that this experimental approach can be used to measure the breakdown toughness of other dielectric materials. Understanding the nature of these initial conducting filaments may enable to tailor the chemistry and microstructure of dielectric materials towards better breakdown resistance. In addition also further theoretical work is needed to determine the electrical breakdown energy release rate as function of the filament length for different sample geometries.

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References


