SIMULATION ANALYSIS OF THE DIELECTRIC POLARIZATION IN LIQUID SILICONE RUBBER

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In this paper, the dielectric behavior of an unprocessed Liquid Silicone Rubber (LSR) sample has been investigated via the approximation of intermittent Polarization and Depolarization Current (PDC) measurements based on Debye's theory. The extended Debye models are compared with regard to the number of required relaxation branches as well as their time constants. It was found that the depolarization of LSR could be modeled predominantly by five distinct Debye models, while the polarization was reconstructed by a span of four to eight, indicating a comparatively high dispersion. The time constants (TC) of these components change in a convergent manner with progressing measurement cycles, which are analyzed in terms of their correspondence to the individual constituents of LSR. Furthermore, it was observed that the polarization current is reduced with increasing number of measurements, which was ascribed to the presence of an additional Debye relaxation, that was not captured due to the relatively short time period of the measurement cycles. A contrast thereof to an analytical description was also presented as a further verification.

1. Introduction

LSR, which stands for a widely used material in electrical insulation systems, is highly regarded for its exceptional electrical, chemical, thermal, and mechanical properties.

With the ongoing shift towards High Voltage Direct Current (HVDC) transmission systems, it has become crucial to have a comprehensive understanding of LSR's electrical behavior under DC stress. Despite extensive research on aging, liquid permeation, and the investigation of mechanical material parameters in the presence and absence of an external electric field, there remains a gap in the modeling of LSR's dielectric behavior.

To address this deficiency, PDC measurements can be employed to capture the time-dependent relaxation behavior of LSR. These measurements reveal discrete polarization phenomena in the material, which can then be extracted using an approximation method based on an extended Debye model. This approach enhances the potential for further interpretations regarding the material's conditions.

This paper aims to provide a comprehensive analysis of the transient dielectric response of LSR using this method.

2. Methodology

2.1. Measurement

The measurements were conducted in a completely automated manner, with the ammeter (Keysight B2981A), HV-Source, and peripheral switching circuitry all interconnected through LabVIEW on a PC. A standard three-electrode setup made of stainless steel was utilized.

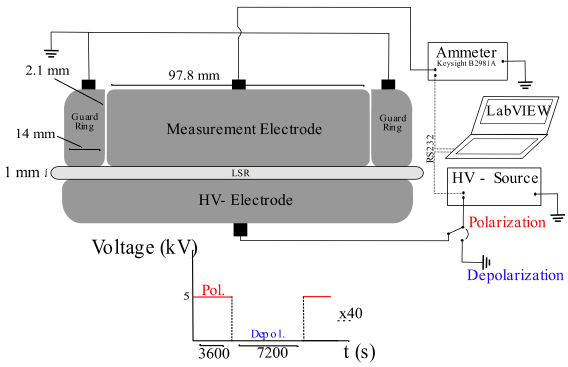

Figure 1: Schematic representation of the automated measurement setup.

The LSR disc used as the material sample had a thickness of 1 mm and was kept under normal atmospheric conditions for a year. It was subjected to a DC voltage of 5 kV for 3600 s, resulting in an average field stress of 5 kV/mm. This polarization phase was always followed by a depolarization phase that lasted twice as long (7200 s). This process was repeated until the LSR reached a stationary[a] condition, resulting in approximately 40 polarization measurements (PM) and 40 depolarization measurements (DM) in total.

2.2. Theory

The ideal polarization of a solid dielectric in the absence of many-body-interactions obeys the exponential relationship of relaxation according to Debye's theory, which is mathematically given as:

$$i(t) \propto A \cdot \mathrm{e}^{-t/\tau} \qquad (1)$$

where i represents the current, t corresponds to time, $A = U/R$ and $\tau = R \cdot C$ represent the magnitude of the response to a voltage waveform of a *Heaviside* function with amplitude U, and the time constant (TC) of the Debye model[b] comprising the resistance R and capacitance C, respectively. In contrast to (1), however, in real dielectrics, the polarization and depolarization current diverge from an ideal behavior in the sense that a partial current that is superposed to the ideal current[c] can be observed over a wide gamut of types of dielectrics [1]. This is generally associated with the *memory-effect* that is inextricable from the ideal current behavior. An all-encompassing mathematical description of the depolarization current, including the peculiarity under consideration, has been posited in [2], [3], under the designation of *Curie-von Schweidler* law long since as:

$$i(t) \propto t^{-n}, n \in (0,1), t \in (t_{\min} \neq 0, t_{\max}) \qquad (2)$$

where n stands for the relaxation exponent in the power law relation of the time dependence within an arbitrary time period demarcated by $t_{\min}$ and $t_{\max}$. Appertaining to its wide-ranged applicability, (2), along with other correlates, has been deemed to be universal to dielectrics, hence dubbed "*Universal Dielectric Response* (UDR)" [1], [4]. (2) can be approximated in form of (1) by

$$i(t) = \sum_{j}^{N} A_j \cdot \mathrm{e}^{-t/\tau_j} \approx C \cdot t^{-n}, \qquad (3)$$

where N is the maximum number of required components for an arbitrary $C \in \mathbb{R}$. A succinct depiction of (3) can be found in Fig. 7.2 of [4]. (3) implies an equivalence of UDR to a weighted linear combination of multiple discrete Debye-type relaxations. This integral behavior is termed as "Distribution of Relaxation Times (DRT)", see Fig. 2, where C_0 and R_0 represent the geometric capacitance and resistance of the LSR respectively.

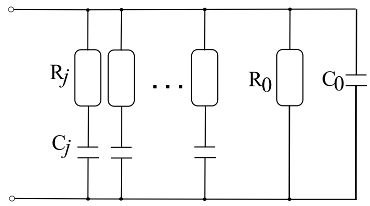

Figure 2: Equivalent circuit diagram of multiple unrelated Debye type relaxations in a dielectric

DRT is underpinned by the plausible interpretation that, there exist numerous discrete polarization phenomena in the dielectric that are heterogeneously distributed and feature different magnitudes and TCs [5] [6]. These shall collectively, and in an isolated manner, lead to the behavior given in (2). The way it is presented in (3), DRT has been disparaged by the pioneers of UDR and other relevant researchers with regards to its purported status as a mere mathematical re-description of (2) without an additional insight into the distinct relaxation mechanisms. In particular, with DRT, there is ostensibly no possibility of discriminating the individual polarization mechanisms with the intention of proving that a hypothetical distribution would apply in different cases. On the other hand, in contrast to the foregoing perspective on the matter, a significant portion of the literature is also devoted to the interpretation that (2) is actually an approximation of the manifested (3) [9] and is indeed inferior to DRT. Notwithstanding the debates on the topic, DRT has been obtained for various materials [6], [7] specifically with regards to the analysis of several aspects of the dielectrics, e.g. determining TCs and the impact of the physical conditions of the materials thereon. However, considering LSR, to our knowledge, there exists only one study [8], that derived an equivalent circuit of LSR aiming at separating the polarization that is engendered by the moisture content in LSR from the polarization of the rest of the composite. It can be stated that, except [8], the corpus on the investigation of the polarization in LSR by the employment of an equivalent circuit is deficient in further work. In this study, we acquired measurement results that obey both (1) and (2), yet deem DRT to be superior to UDR in terms of its generalizability, and consequently deploy DRT to explicate the dielectric behavior of LSR.

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[a] by "stationary" we imply the condition in which the current could be reproduced without any significant difference to a previous measurement, see also Fig. 3b

[b] from now on we refer to a Debye model with "component", representing a single branch in Fig. 2.

[c] one that comports with an isotropic dielectric constant ε and conductivity κ of the material

2.3. Modeling

Pursuant to the explanation in [7], we utilize the Algorithm 1 to obtain the components from PDC measurements of an LSR sample[d].

Algorithm 1 Approximation Procedure

Set $\eta_1 := \eta \rightarrow$ number of data points

For $k = 1, 2, 3, \ldots$

  For $m \in \{1, \ldots, \eta_k - 1\}$

    Solve for $A_k^m > 0, \tau_k^m > 0$ **with**

$$\text{(a)}\quad \begin{aligned} A_k^m \cdot \exp(-t_{\eta_k} / \tau_k^m) &= i(t_{\eta_k}) - \sum_{j=1}^{k-1} A_j \cdot \exp(-t_{\eta_k} / \tau_j) \\ A_k^m \cdot \exp(-t_m / \tau_k^m) &= i(t_m) - \sum_{j=1}^{k-1} A_j \cdot \exp(-t_m / \tau_j) \end{aligned}$$

  Set $m_k^* := \underset{m \in \{1, \ldots, \eta_k - 1\}}{\operatorname{argmin}} A_k^m$

  Set $\tau_k := \tau_k^{m_k^*}, A_k := A^{m_k^*}$

  If $m_k^* > 2 : \rightarrow$ at least 2 data points are required

    Set $\eta_{k+1} := m_k^* - 1$

  Else:

    Set $N := k$

    Exit

Briefly, starting with the last available data point, Algorithm 1 traverses all of the data points, seeking an additional data point, with which the solution of the given equations in Algorithm 1-(a) results in a pair (A_k, τ_k), where A_k is minimized. Subsequent to every extracted pair, it shrinks the data upto the index m_k^*, which corresponds to the second data point required to find the previous pair, continuing with the search of the next pair until the data has been exhausted.

3. Results

3.1. Current Measurements

The results of the subsequent PDC measurements on LSR along with the corresponding approximations estimated by Algorithm 1 are illustrated in Fig. 3a and 3b. It can be observed that, in every consecutive measurement pair, the overall current in the polarization (depolarization) phase is gradually lowered (increased) without any exceptions up to the 36th measurement.

Qualitatively, the relaxation behavior observed aligns well with the findings in [9], echoing an anticipated outcome according to [10], with regards to the link between consecutive polarization and electrical history of the material.

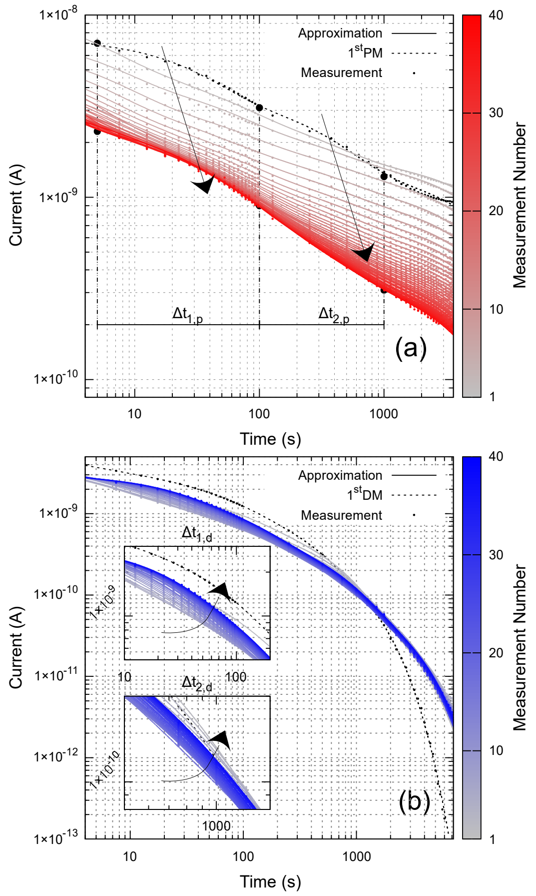

Figure 3: Subsequent polarization (a) and depolarization (b) measurements, along with the individual approximations thereof.

Additionally, as a prelude to the Debye models in section 3.2, four distinct regions in the measurements were found to be of interest to the interpretation. These are $\Delta t_1 := (5\,\mathrm{s}, 100\,\mathrm{s})$ and $\Delta t_2 := (100\,\mathrm{s}, \sim 1000\,\mathrm{s})$, emphasized either through additional tics on time axis as in Fig. 3a or as a zoom-out as in Fig. 3b. Within the hitherto mentioned time periods, both type of measurements converge to a definite current trend, where distinguishable transitions in the slope of the current can be seen, pointing to distinct relaxation mechanisms that become more discernible as the measurement cycles progress.

3.2. Debye Models

Prior to the illustration of the TCs of individual components, we shall evaluate the frequency distribution of the number of required components, depicted in Fig. 4. By utilizing Algorithm 1, we obtained 34 models out of a total of 38 DMs that rendered the LSR

[d]without the exclusion of other dielectrics, as Algorithm 1 could be successfully tested on PDC measurements of oil-impregnated pressboard and mineral oil as well

to contain five components for the loading configuration studied here.

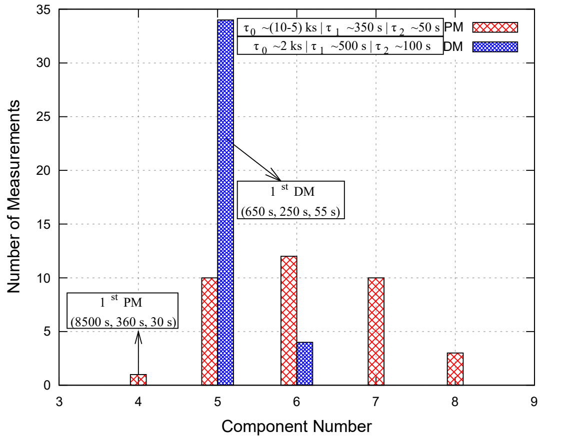

Figure 4: Frequency distribution of the number of components. The components with the 3 greatest TCs independent of the component number are explicitly given accompanying the legends

The rest required only an additional one. In contrast to that, PMs showed a comparatively high degree of variation in the number of components, ranging from four to eight, moderately consistent with the findings in [8], especially considering the fact that the LSR sample used in this study did not undergo any appreciable conditioning or purification process. Here, the case with six components can be singled-out, albeit only marginally, as the most frequent occurrence.

Considering the individual TCs, it can be stated that the initial measurements had slightly different TCs than those of the succeeding ones. Furthermore, precluding the initial measurements and neglecting the minute variations in TCs, the greatest three TCs were virtually identical throughout the measurement series independently of the total component number, i.e. where applicable, additional TCs were required merely to adapt the approximation to the current curve within the first 10 s. Consequently, we surmise that, solely the analysis of the models with five components should sufficiently enable us to interpret the behavior in $\Delta t_{1,\mathrm{d}}$, $\Delta t_{2,\mathrm{d}}$, $\Delta t_{1,\mathrm{p}}$ and $\Delta t_{2,\mathrm{p}}$. The relevant TCs are shown in Table 1. TCs seem to shift with the measurement number. Also note that the shift in TCs are for both measurement types in the same direction. For instance, both $\tau_{0,\mathrm{d}}$ and $\tau_{0,\mathrm{p}}$ decrease over the measurements, which potentially indicates that the identical relaxation processes take place in PMs and DMs, though with distinct absolute TCs, discussed in section 4.

Table 1: Shift of TCs with repetitive (↓) loading.

TC[ks]	τ_0	τ_1	τ_2	τ_3	τ_4
PM 1st ↓ 36th	10 ↓ 5	0.36 ↓ 0.33	0.03 ↓ 0.07	0.01 ↓ 0.02	$\approx 5\cdot 10^{-3}$
DM 1st ↓ 36th	2.2 ↓ 2	0.54 ↓ 0.47	0.06 ↓ 0.09	0.01 ↓ 0.024	$\approx 5\cdot 10^{-3}$

4. Discussion

4.1. Shift of TCs with repetitive loading:

In advance of the physical interpretation of the TCs, the most influential components within Δt_1 and Δt_2 should be discerned[e] and the effect of their alteration on current should be analyzed. To that purpose, Rs and Cs are modified in SPICE models of 1st PM and 1st DM, see Fig. 5.

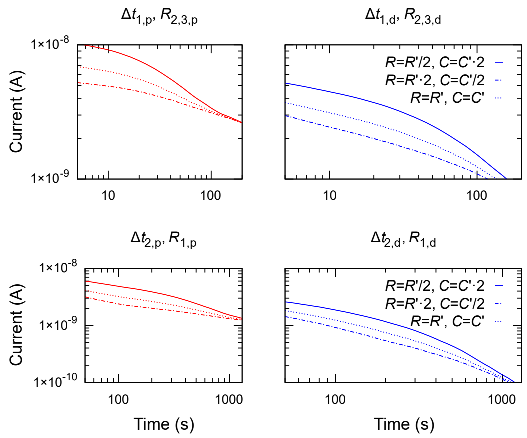

Figure 5: Variation of Rs and Cs, in analogy to [7].

Consistent with the given TCs in Table 1, $R_{2,3,\mathrm{p}}$ and $R_{2,3,\mathrm{d}}$ are the most dominant within Δt_1, whereas in Δt_2 the same can be stated for the modification of $R_{1,\mathrm{p}}$ and $R_{1,\mathrm{d}}$. Consequently, analysis of these over the measurement cycles should sufficiently facilitate the investigation of the correlating relaxation phenomena, which are shown in Fig. 6 in normalized values[f]. Ignoring the $R_{0,\mathrm{p}}$ for the following discussion, from Fig. 6 it can be recognized that $R_{2,3,\mathrm{p}}$ plateau after the 10th measurement, and taking heed of the fact that $\tau_{2,3,\mathrm{p,d}}$ rise according to Table 1, we reason that the relatively low TCs

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[e] τ_4 is neglected, since at $t = 10\,\mathrm{s}$, this component will have lost ~86% of its current contribution.

[f] Obviously all of the Rs increase with the measurement number so that only the relative differences contain the essential information.

turn capable of storing further charge after every measurement cycle, simultaneously shifting the aforementioned “bumps” slightly to longer time periods, see also Fig. 5.

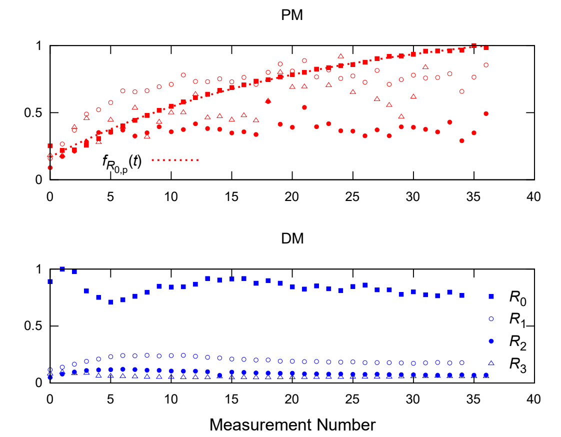

Figure 6: Rs over the measurement number. Exponential fit in form of $f_{R_{0,p}}(t)=c_1-c_2\cdot\exp(-t/\tau)$, *where* $\tau=\tau_{R_{0,p}}\approx 240.256\,\mathrm{ks}$ *is valid.*

In a similar vein, beginning from the stagnation of $R_{2,3,\mathrm{p}}$, a slight decrease of $R_{2,3,\mathrm{d}}$ set in, resulting in equivalently higher current in $\Delta t_{1,\mathrm{d}}$. With above assertions, following can be postulated:

In general, as is also suggested in [7], it is expected for the short TCs (τ_2,τ_3) to correlate with the dielectric behavior of the liquid part of the LSR, which stems from the higher mobility of the charge carriers therein. In contrast to that, larger TCs are presumably originated from the relaxation processes that possess higher “inertia”, e.g. *Maxwell-Wagner* polarization. Alas, here, it is not possible to differentiate between these solely with the obtained TCs as there is no quantitative comparison to another material sample in terms of the TCs. Still, if $\tau_{2,3,\mathrm{p}}$ were to be related to the dielectric relaxation within the liquid constituent, the growth of $R_{2,3,\mathrm{p}}$ could be deemed attributable to the sweep-out of the free charge carriers within the liquid, which partially become trapped at the interface regions of the polymere matrix. Within the subsequent depolarization, their release is facilitated as the measurement number rises, i.e. $R_{2,3,\mathrm{d}}$ lessen. Analogous to $R_{2,3,\mathrm{p}}$, $R_{1,\mathrm{p,d}}$ too level off after ~10th measurement. However, conversely, $\tau_{1,\mathrm{p,d}}$, and with that $C_{1,\mathrm{p,d}}$, decline with the measurement number, which can be hypothesized to be ensued from the saturation of the corresponding relaxation branches, unable to hold onto any extra charge. Assessing the Rs in DMs in isolation, and realizing that the modification of $\tau_{0,\mathrm{d}}$ is exceedingly less than that of $\tau_{0,\mathrm{p}}$, it can be stated that in DMs we obtain an equivalent circuit of LSR that is starkly less variant to measurement number unlike that in PMs.

4.2. Gradual reduction in PMs with repetitive loading:

In the last section, the modification of $R_{0,\mathrm{p}}$ has been glossed over, which assumably partook in the succesive reduction of PMs, see Fig. 3a. To reappraise the drop rate in PMs and relate it to $R_{0,\mathrm{p}}$, total charge from PMs and DMs are plotted in Fig. 7[9].

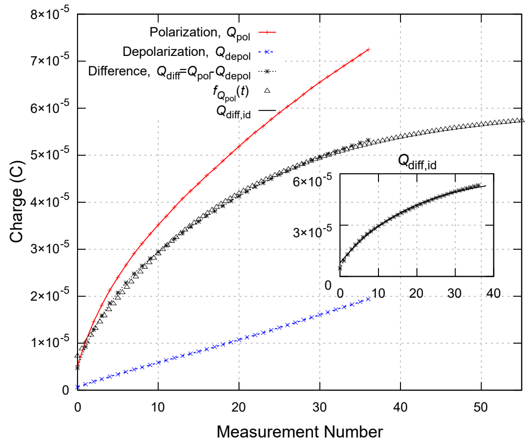

Figure 7: Total charge inserted and extracted in PMs and DMs respectively. Exponential fit in form of $f_{Q_{pol}}(t)=c_1-c_2\cdot\exp(-t/\tau)$, *where* $\tau=\tau_{Q_{diff}}\approx 204.18\,\mathrm{ks}$ *is valid.*

It is worth noting that Q_{pol} is converging exponentially toward its long-time limit, which points to the presence of a component with a vastly greater TC compared to those given in Table 1. Ideally, in a repetitively loaded component, Q_{diff} can be analytically calculated as:

$$Q_{\mathrm{diff,id}}(k,\Delta t_{\mathrm{p}},\Delta t_{\mathrm{d}})=U\cdot C\sum_{k=1}(f_d\cdot f_p)^k\cdot(1/f_p-1)$$
$$f_p=\exp(-\Delta t_{\mathrm{p}}/\tau_p), f_d=\exp(-\Delta t_{\mathrm{d}}/\tau_d), \qquad (4)$$

where k is the number of measurement cycles carried out as pairs of PM and DM of durations Δt_{p} and Δt_{d}, respectively. Reconstruction of measured Q_{diff} by (4) with $\tau_d=\tau_p=\tau_{Q_{\mathrm{diff}}}$ represents a strong evidence that, apart from the explanations in the previous section, the LSR sample behaved quite linearly with repetitive loading, which is reaffirmed in Fig. 7.

[9] To calculate the charge, the conduction current from a long-time PM was subtracted from all of the PMs.

With that we suggest that the reduction in the PMs was indeed the result of the charging of the corresponding linear component. Considering that $\tau_{R_{0,\mathrm{p}}} \approx \tau_{Q_{\mathrm{diff}}}$, $R_{0,\mathrm{p}}$ might be partially involved within the putative component, which could have been captured erroneously by Algorithm 1 due to the comparatively short duration of the PMs.

5. Conclusion

In this contribution the dielectric relaxation of an LSR sample has been evaluated via PDC measurements and their approximation based on an extended Debye model. It was found that the LSR sample could be modeled by 5 components in depolarization, and 4 to 8 components in polarization. The TCs of the components seem to vary as the material is repeatedly loaded.

Here, two aspects with regard to the measurements were analyzed. Firstly, an effort has been made to distinguish the constituents of the composite with regard to their relaxation mechanisms. Relying on the change in short TCs and their Rs, it is proposed that the charge carriers in the liquid part of LSR could have been swept out and trapped in the polymere matrix, leading to greater apparent R of the corresponding component, as these will be absent from then on and will cease their contribution to the current. Furthermore, a second component with a comparatively longer TC was detected, which, akin to the liquid relaxation above, showed a stagnation of its resistance. However, in contrast to an increase in the capacitance of components from the liquid, this one indicated a diminished charge storage capability, which was interpreted to be a saturation of the putative component. To validate the above interpretation, it is necessary to replicate the present study with an LSR sample that has been dried under vacuum, so that the volatile constituents are vaporized, eliminating the correlating components and simulatenously allowing to distinguish between these. Secondly, a gradual reduction in the PMs was observed throughout the measurements. This was ascribed to the presence of an additional component with a significantly greater TC, which was evinced by the fact that an analytical description of the stored charge in a component could reproduce the measured accumulated charge within LSR. From this vantage point, we can state that LSR behaves exceptionally linearly for the investigated electric field strength level in this study. The accuracy of the estimation of the TC for the component in question can be evaluated by a long-time measurement of an LSR sample that has been kept under identical conditions as the one investigated in this study.

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