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Effect of Humidity on Lifetime and Dielectric Properties of PET Film

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Abstract-Due to recent developments in semiconductor switching technology, the associated insulation systems are exposed to higher pulse voltage amplitudes and frequencies as well as shorter rise times, which might accelerate material degradation. Often, the insulation is also subjected to humidity changes which affect the dielectric properties and possibly the insulation lifetime. This motivates the study of humidity effects under DC-biased pulse-width modulated (PWM) voltage stress. The aim of this contribution is to provide an experimental basis as well as explanations for humidity-induced variations in the dielectric properties and changed lifetime behavior of PET film. Throughout all experiments, the relative humidity (RH) of air is varied. The lifetime is evaluated as time-to-failure (TTF) by applying DC voltages with and without superimposed PWM voltages below and above the partial discharge inception voltage (PDIV). In order to quantify changes in the dielectric properties, the complex dielectric permittivity is evaluated by means of broadband dielectric spectroscopy (BDS). The water absorption is measured gravimetrically. Although the observed relative water absorption of the PET specimens is always below 0.37 %, the lifetime is drastically decreased by more humid conditions under DC stress (below PDIV). Above PDIV, PD erosion dominates at low RH, whereas DC breakdown processes become more significant at high RH. The BDS results reveal humidityinduced changes in the relaxation and conduction behavior. Finally, the breakdown mechanisms, which are altered by water absorption, are discussed in terms of thermal and space-chargedriven breakdown.

I. INTRODUCTION

The development of wide band gap semiconductor (SiC, GaN) technologies paved the way for new power switching applications [1]. This implies higher pulse voltage amplitudes and switching frequencies as well as increased switching speeds (> 50 kV/ μ s) [2], which might accelerate material degradation. Often, the insulation system is also subjected to environmental influences which affect the dielectric properties and possibly the material's lifetime. This contribution focuses on the humidity effects on the dielectric properties and lifetime of polyethylene terephthalate (PET) films, which have been used as electrical insulation material for many years.

Four main risk factors have been identified [3] for insulation systems in medium voltage (MV) converter systems. First of all, *partial discharge (PD)* repetition rate increases with switching frequency and leads to material erosion [4-6]. Furthermore, voltages with a broad spectral content may generate resonances or standing wave phenomena (e.g. at motor windings) [5]. These *systemic overvoltages* can in turn incept PDs [7]. Aging in the absence of PDs has also been reported [6,8,9]. One explanation is given in terms of *excessive dielectric heating* (due to the alternating electric field) which can initiate a breakdown by means of material degradation or thermal breakdown. Polar materials are found to be more vulnerable to this dielectric heating (compared to non-polar materials) due to the strong orientation polarization present in the material [10]. In addition, *space charges* can build up under DC fields and possibly lead to electromechanical aging (Coulombic forces on space charges) if a highly dynamic field is superimposed [11].

In a humid atmosphere, water diffuses through the amorphous regions of a semicrystalline polymer [12]. The water molecules can either attach themselves to polymer groups (e.g. hydroxyl group -OH) or exist as free/clustered water in the amorphous volumes or in microvoids [13-15]. With respect to the material's lifetime, humidity influences on surface as well as on bulk properties need to be considered. It is reported that surface conductivity increases with the level of relative humidity (RH) [16], thus spreading the PDs over a larger area which increases lifetime [17]. This effect is present especially for low-hygroscopic materials [18]. However, above a critical value RH_c, charges (deposited after a discharge) can be removed totally from the surface, consequently removing their shielding effect. This, in turn, decreases the lifetime of the material [19]. In the absence of PDs, the breakdown behavior might also be altered by humidity. For instance, the polar water molecules can lead to dissociation of impurities inside the polymer which increases DC conductivity and, as a consequence, might initiate thermal breakdown [14,20]. Moreover, these humidity-induced charge carriers can be trapped (between the amorphous and crystalline phases), leading to buildup of (more) space charges under an applied DC field [21]. The absorbed water might also change the internal field distribution, which might result in a (non-)uniformly reduction of the effective specimen thickness. This can drastically decrease the breakdown strength [22].

The dielectric properties of a material are represented by its dielectric spectrum. Previous studies showed a dielectric permittivity increase (polar nature of water) and a dissipation factor increase (water molecules bound to the –OH group of the PET chain ends) with increasing humidity for PET [23,24]. In order to investigate further the breakdown processes (under DC with and without rectangular pulse voltage) in PET under humid conditions, this contribution provides a lifetime

study both in the presence and in the absence of PDs. Moreover, broadband dielectric spectroscopy (BDS) is used to relate the humidity-induced changes of the dielectric properties to the lifetime measurements. The present contribution follows a previous study [25], wherein the dielectric properties of PET under temperature and humidity variations are examined.

II. EXPERIMENTAL

A. Materials and Water Absorption

Measurements are conducted on semicrystalline, biaxially oriented PET film (DuPont Melinex[®] S) with a thickness of $23 \pm 0.5 \,\mu$ m. The foil is cut into circles of 50 mm and 30 mm diameter for lifetime and BDS measurements, respectively. The glass transition temperature T_g is around 78..80°C [26,27]. In order to quantify the water absorption of the investigated film, gravimetric analysis is used. The detailed study is reported in [25] and reveals a linear dependence of the absorbed water and the RH level. At RH = 80 %, a relative weight gain of 0.37 % is reached at 23 ± 1 °C and the good accordance with Fick's law of diffusion allows the calculation of the diffusion constant $D_{\text{PET}} \approx 3.9 \cdot 10^{-9} \,\text{cm}^2/\text{s}.$

B. Inverter Stress Simulator for Lifetime Analysis

The schematic for the lifetime measurement setup is illustrated in Fig. 1 (a). Details of the construction and validation of this setup are found in [3]. The pulse-width modulated (PWM) rectangular pulse voltage is generated by means of a MOSFET half-bridge S1/S2 which is fed by a DC source U_p with a DC-bus capacitor $C_1 = 1 \mu F$. These unipolar pulses are superimposed through the coupling capacitor $C_2 = 2.7 \text{ nF}$ (thus becoming bipolar) onto the DC voltage U_{DC} , which is generated by an AC voltage source U_{AC} (50 Hz) and a simple rectifying circuit. Decoupling between DC and pulse voltage is provided by $R_3 = 1 \text{ M}\Omega$. In case of a specimen breakdown, the resistors $R_1 = 220 \Omega$ and $R_2 = 50 \Omega$ as well as the Diodes D_1/D_2 protect the half-bridge. A current transformer (CT) serves for breakdown detection and initiates shutdown of the voltage sources as well as blocking of the half-bridge.

The test cell with electrode-specimen configuration is shown in Fig. 1 (b). In order to control the RH level at room temperature, a dry air flow is separated into two branches, of which one passes through a distilled water bath. The humidity is changed by adjusting the volume flow of each branch according to the measured RH value in the mixing chamber.

For lifetime tests, the specimens are placed between a halfsphere (radius 5 mm) and a plate electrode (diameter 10 mm, fillet radius 1.5 mm) made of aluminum. The defined contact force (by means of a spring) leads to an indentation of the grounded electrode in the PET foil over a diameter of 0.16 mm and a maximum depth of around 1.2 μ m (including electrostatic force between electrodes) [3]. After each breakdown, the specimens and electrodes are replaced and the electrodes are polished to eliminate influences by residuals of the breakdown.



Fig. 1: a) Schematic of inverter stress simulator with exemplary voltage waveforms; b) test cell with temperature and humidity control circuits.

In order to compare effects in the presence and absence of (external) PDs, the partial discharge inception voltage (PDIV) is measured via a UV sensitive camera. This results in a value $U_{\text{PDIV}} = 1.0..1.1 \text{ kV}$ for the pulse voltage (peak-to-peak). Färber et al. found the PD repetition rate due to DC stressing to be negligibly small (<1 s⁻¹) [3]. The overshoot during the pulse voltage rise is observed to be <5%. Voltage measurements are performed by means of a LeCroy PPE 20kV/100MHz HV probe.

The test voltage form is chosen in a way that the peak voltage at the device under test $U_{\text{DUT,p}}$ is constant and U_{DC} (i.e. averaged voltage over one period) does not vary significantly:

$$U_{\text{DUT,p}} = U_{\text{DC}} + U_{\text{p}} \cdot (1-D) = 10.25 \text{ kV}, \text{ with } D = 0.99$$
(1)

In Tab. 1, the test conditions (voltages, switching frequency f_s , duty cycle *D*, pulse rise time τ_r) used in this work are given.

	U _{DC} in kV	U _p in kV	<i>f</i> _s in kHz	D	τ _r in ns	T _{room} in °C
no PD	10.25	-	-	-	-	23 ± 1
	10.2425	0.75	4	0.99	40	
PD	10.235	1.5				

Table 1: Test parameter values for lifetime measurements on PET film

C. Broadband Dielectric Spectroscopy (BDS)

Dipolar as well as charge migration effects in PET (when exposed to an external electric field) can be described by the frequency-dependent complex relative permittivity

$$\varepsilon_{\rm r}^* = \varepsilon_{\rm r}' - {\rm j}\varepsilon_{\rm r}''. \tag{2}$$

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The real part ε'_r is a measure of the polarizability of a material. The imaginary part ε''_r describes the material's dielectric losses and consists in general of the DC conductivity loss component $\sigma_{DC}/(2\pi f \varepsilon_0)$ as well as the polarization losses $\varepsilon''_{r,pol}$. These are used to evaluate the real part of the complex conductivity [28]:

$$\sigma' = \omega \varepsilon_0 \varepsilon_r'' = \sigma_{\rm DC} + \omega \varepsilon_0 \varepsilon_{\rm r,pol}'' \tag{3}$$

The construction and working principle of the used BDS setup in this work are described in [25,29]. PET specimens with metallized (silver painted) guarded electrodes (3 mm diameter) are used. Prior to each measurement, the specimens are dried at 50 °C and stored under the desired RH atmosphere for > 1 day. This is sufficient to reach saturated water absorption ($t_{sat} \approx 11.5$ min). The BDS measurements in this work are performed at 10 °C in a frequency range $10^{-2}..10^{5}$ Hz for different RH levels.

III. RESULTS

A. Time-to-Failure (TTF) Measurements

The lifetime of the stressed specimens is evaluated in terms of time-to-failure (TTF) measurements and fitting the data according to a two-parameter Weibull distribution (for details see [3]). Each data point in Fig. 2 represents the median (with 95 % confidence interval) out of ten TTF measurements.

With increasing RH level (i.e. linearly increasing amount of absorbed water, see [25]), the TTF decreases for all test conditions. In the absence of PDs, the TTF is identical for the two cases of DC with and without PWM pulse voltage stress. Above PDIV ($U_{PDIV} = 1.0..1.1$ kV), the low-humidity lifetime is much lower than without PDs. However, for higher RH levels, the decrease of TTF decelerates and almost the same (short) lifetime is reached for the highest measured RH value.

B. Broadband Dielectric Spectroscopy (BDS) Measurements

The real part of the complex relative permittivity ε'_r , shown in Fig. 3, increases approximately linearly with the RH level. At high frequencies, the imaginary part ε''_r exhibits a β relaxation peak at dry as well as at humid conditions for PET, which increases slightly in magnitude with higher water absorption level. For RH \geq 33 %, an additional loss mechanism appears.

Its peak frequency (i.e. reciprocal relaxation time) increases with increasing humidity. Furthermore, in the lowest investigated frequency range (< 1 Hz), a significant increase of ε_r'' with absorbed water emerges. It should be mentioned that the reported changes in Fig. 3 are reversible after drying at 50 °C for > 1 day and cooling down again to 10 °C.

IV. DISCUSSION

A. Influence of RH Level on Lifetime of PET

The saturated water absorption during the TTF measurements presented in this work is always below 0.37 % (relative weight gain). Despite this low amount of water, the lifetime under all investigated electrical stress conditions decreases drastically (almost exponentially) with increasing RH level, see Fig. 2. Above PDIV, surface erosion by surface discharges is observed (see study in [3]), thus leading to a decrease in lifetime compared with the PD-free regime. No TTF increase is observed for rising RH level if PDs are present. This indicates that a surface effect of humidity, i.e. spreading of PD over larger area due to increased surface conductivity as found in [17,19,30], plays only a minor role (if at all). Moreover, above PDIV, no accelerated lifetime decrease with increasing humidity (due to possible removal of shielding charges from the surface at high RH levels as in [19]) is visible in Fig. 2. Thus, mainly volume effects are assumed to determine the humidity-dependent breakdown behavior of the investigated PET film.



Fig. 2: Dependence of measured TTF on RH level (saturated water absorption) for PET foil $(23 \pm 0.5 \ \mu m$ thickness) at $23 \pm 1 \ ^{\circ}C$. The inset plot represents the same data in log-linear scale.



Fig. 3: Frequency variation of the real (top) and imaginary part (bottom) of the complex relative permittivity at 10 $^{\circ}$ C for PET at different RH levels (saturated water absorption). The arrows indicate the mid-frequency loss peaks.

Below PDIV, no additional breakdown processes are introduced by the superimposed PWM pulse stress. This means that there is no accelerated breakdown contribution of water molecules reacting to the alternating field (at least for $f_s = 4$ kHz). Consequently, in the PD-free regime, the DC breakdown mechanism causes the breakdown in PET and can be influenced by absorbed water. The difference between the PD-free and the PD regime decreases with increasing RH level. This means that PD erosion (as long as present) dominates and accelerates the breakdown in the low-humidity range, whereas the DC breakdown mechanism becomes more significant in the high-humidity range. Finally, at RH = 75 %, a difference between PD-free and PD regime is no longer apparent, because the DC mechanism initiates the breakdown at least as fast as the PD erosion. A better understanding of the RH influence(s) on lifetime, however, requires additional knowledge about the hereby introduced changes of the dielectric properties, which will be given in the following section.

B. Influence of RH Level on Dielectric Properties of PET

The linear ε'_r increase with RH level in Fig. 3 results from the higher polar content due to the absorbed (polar) water molecules [31]. It was proposed in an earlier study [25] that water exists in PET film in (at least) two different forms. On the one hand, water molecules are bound to the terminal –OH groups of the PET chain, thus changing the β relaxation [23,24]. This is visible in the high-frequency range of ε''_r in Fig. 3. On the other hand, free or clustered water is likely to exist in the free volume of the polymer (e.g. in amorphous regions, at grain boundaries or in small voids).

An indication for clustered water inside the polymer can be given by the mid-frequency relaxation mechanism which appears in Fig. 3 only for RH \geq 33 %. Its peak is shifted to higher frequencies (i.e. lower relaxation times) with increasing humidity. In principle, such a behavior can be attributed to a plasticization effect by the water, thus lowering T_g [13,32]. Since a T_g decrease was found by Jabarin et al. to be only about $\Delta T_g \approx 16$ °C at RH = 80 % (for $T_{g,\text{PET,dry}} \approx 78$ °C) [26] and the BDS measurements in this work are performed at 10 °C, plasticization is excluded as a reason for the observed peak shift. Another explanation is given in terms of (loosely bound) water layers which exhibit a conductivity increase with higher level of water absorption [33]. These semi-conductive regions can be seen as inhomogeneities inside the polymer, thus distorting the internal electric field distribution by means of interfacial polarization.

In the low-frequency range, the ε_r'' increase with the RH level in Fig. 3 is attributed to a growing contribution of DC conductivity in the dielectric losses. By evaluating the BDS data by means of (3), Fig. 4 is generated which shows the DC conductivity increase in PET with rising RH level at low frequencies. The absence of a σ' plateau indicates that the macroscopic DC conductivity does not yet dominate over polarization mechanisms in the investigated frequency range. Combined with the assumption of localized water layers, it is more likely that DC conductivity increases strongly locally with absorbed water. At this point, it should be noted that even at the lowest frequency of 0.01 Hz, the sinusoidal excitation voltage changes polarity every 50 s, whereas the shortest TTF is in the range of 4..5 min. Consequently, the BDS measurements only slightly reflect the DC conductivity behavior in the critical time range, but indicate the presence of more involved mobile charge carriers by water absorption. These can result from the water molecules themselves or from dissociation of ionic molecules into ionic charge carriers by the polar nature of water [20,28].



Fig. 4: Frequency variation of the real part of the complex conductivity for different relative humidity levels according to (3) and same data as in Fig. 3.

C. Possible Reasons for RH-induced Lifetime Decrease

The lifetime of the investigated PET specimens is not influenced by any AC effects (at least under the test conditions in Tab. 1) if no PDs are present, which is in accordance with the findings in [3]. This is supported by the low increase in the β relaxation in the high-frequency range (Fig. 2) which is not sufficient to increase dielectric losses over a critical thermal limit. Therefore, the following discussion focuses on the decreased lifetime (or TTF) by means of humidity under DC stress. One explanation is given in terms of a thermal breakdown due to dissociation of impurities by polar water molecules which leads to a DC conductivity increase that was detected in Fig. 3 and Fig. 4. This heats up the polymer locally which further increases DC conductivity and finally initiates the breakdown [20]. Furthermore, space charge accumulation and thermally activated hopping at the crystalline-amorphous interfaces can be altered by absorbed water and thus contribute to the breakdown in PET [21,34,35]. Water absorption is also likely to lead to a non-uniform water distribution inside the dielectric. This, in turn, distorts the local electric field distribution and consequently lowers the breakdown strength [22], which is in accordance to the model of semi-conductive water layers proposed in this contribution.

In principle, either space charge evolution (leading to inhomogeneous field distribution) or heating processes might be in the order of the observed TTF. Thus, a combination of thermal and space-charge-driven breakdown, which both are changed by water absorption, is conceivable.

V. CONCLUSION

The lifetime of PET film is evaluated in terms of time-tofailure (TTF) measurements under DC stress with and without superimposed rectangular pulse voltages. The influences of relative humidity (RH) on the lifetime as well as on the dielectric properties are investigated. The following conclusions are drawn from the obtained results:

- TTF decreases almost exponentially with increasing RH level despite the low amount of absorbed water (0.37 % relative weight gain at RH = 80 %).
- TTF is changed by the RH influences on the electrical bulk properties of PET since no effect on the surface properties (even in the presence of PDs) is observed.
- Below PDIV, superimposed rectangular pulses do not introduce additional breakdown processes due to the low

RH-induced increase in dielectric losses at high frequencies. Accordingly, the DC breakdown mechanism is responsible for the breakdown and can be influenced by absorbed water.

- PD erosion (if present) dominates breakdown in the low-• humidity range, whereas the DC breakdown mechanism becomes more significant at high humidity (initiating the breakdown faster than PD erosion).
- Absorbed water leads to a slight DC conductivity increase and exists in PET as bound water (enhancing the β relaxation) and free/clustered water (inducing an additional loss mechanism). The latter creates semiconductive regions, thus distorting the internal electric fields. Accordingly, thermal and/or space-charge-driven breakdown (both changing with RH level) are likely to cause the observed lifetime behavior.

As a consequence, the present contribution demonstrates the drastic (almost exponential) decrease in TTF for PET film under linearly increasing RH levels (of air). A disregard of these effects would thus result in considerable errors when designing polymeric insulation systems. The measurements in this work, however, are performed with field strengths which are higher than in real applications in order to accelerate lifetime studies. Accordingly, it needs to be examined whether these effects still occur under more application-oriented, lower field strengths.

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