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Published Version:
Suraci, S.V., Fabiani, D. (2021). Aging assessment of insulating materials through broadband dielectric measurements: the appropriate frequency choice. New York : IEEE [10.1109/CEIDP50766.2021.9705347].

Availability:
This version is available at: https://hdl.handle.net/11585/876993 since: 2022-03-02

Published:

DOI: http://doi.org/10.1109/CEIDP50766.2021.9705347

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(Article begins on next page)

10 October 2026

This is the final peer-reviewed accepted manuscript of:

S. V. Suraci and D. Fabiani

Aging assessment of insulating materials through broadband dielectric measurements: the appropriate frequency choice

IN:

2021 IEEE Conference on Electrical Insulation and Dielectric Phenomena (CEIDP), 2021, pp. 32-35

The final published version is available online at:
https://doi.org/10.1109/CEIDP50766.2021.9705347

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Aging assessment of insulating materials through broadband dielectric measurements: the appropriate frequency choice

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***Abstract*- In this article the dielectric response of differently filled insulating materials is studied. The contribution of polymer additives showed to influence both the dielectric spectrum and conductivity values of the considered materials. Among the different analyzed frequencies, it has been concluded that the highest frequency region (e.g. 100 kHz), related to the dipolar polarization, is affected significantly by additives and it depicts a monotonous increase with aging time, hence it could be suitable as an aging marker for cable nondestructive diagnostics.**

I. Introduction

In the last decades innovative diagnostic methods for evaluating the aging state of insulating materials are gaining more and more interest in the electrical engineering field. Up to now, dozens of different techniques ranging from chemical (microscale) and mechanical (macroscale) ones have been successfully developed. According to [1] an ideal condition monitoring technique for electrical insulations should be: nondestructive, easily replicable with low error dispersion, performable in situ and it must be referred to the bulk of the insulation, hence it should be not related to local defects or superficial damages. Up to now, the most common techniques for insulation condition monitoring like e.g., thermo-chemical analyses and mechanical tests are usually destructive and strictly local measurements. Moreover, they require lab equipment, so that they cannot be performed in situ.

The most promising techniques for insulation diagnostic are based on electrical tests, particularly dielectric spectroscopy and conductivity measurements. These techniques meet the demands described above as they are non-destructive techniques, performable in situ and they provide information on the insulation bulk. Electrical properties are usually measured under DC, low frequencies or at power frequencies (50 or 60 Hz). On the other side, broadband dielectric spectroscopy allows the definition of e.g., complex permittivity throughout a wide range of frequencies e.g., between 10^{-2}-10^{6} Hz. At present, very few works consider the impact of the frequency on the proposed aging marker [2]–[5]. Hence, the choice of the frequency which best correlates the aging evolution has still to be properly defined.

This article aims at providing a comparison among the results coming from conductivity and dielectric spectroscopy measurements on radio-chemically aged insulating materials. In particular, the choice of the most appropriate frequency for the material aging evaluation is presented and discussed.

The project leading to this application has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 755183.

II. Materials and Methods

A. Materials

In order to investigate the additive impact on the investigated electrical properties, XLPE-based materials with increasing complexity in fillers e.g., antioxidants and flame retardants are employed. Specimens are obtained in form of tapes with thickness of ~500μm. Due to the roughness of the surface, a Ø (2 cm) circle gold electrode has been deposited through cold-sputtering to ensure good contact between electrode and tested material. In the case of conductivity measurements, electrode deposition is done according to geometries suggested in ASTM D257-14. Materials compositions are reported in Table 1.

TABLE I. Specification and Chemical Composition of Samples

Sample number	Sample composition
#1	Silane crosslinked polyethylene matrix (XLPE)
#2	Silane crosslinked polyethylene matrix (XLPE) + 50phr flame retardants (ATH) + both antioxidants

B. Accelerated aging

Accelerated aging was performed under radiative environments, employing a ^{60}Co γ-ray source. Aging conditions are summarized in Table 2.

TABLE II. Accelerated Aging Conditions

Aging type	Aging properties		
	Dose rate (Gy/h)	Sampling time (h)	Max. absorbed dose (kGy)
Low	7	3456	81
Medium	77	864	333
High	400	167	334

C. Dielectric spectroscopy measurements

Electrical response of samples as a function of frequency is investigated by means of the dielectric spectroscopy technique,

allowing the evaluation of the complex permittivity as a function of frequency.

Dielectric spectra were obtained through a Novocontrol Alpha Dielectric Analyzer v2.2 with applied voltage of 3 V_{rms} at room temperature. The frequency range analyzed is 10^{-2}-10^{6} Hz.

D. Conductivity measurements

Insulation conductivity was measured according to the ASTM D257-14 standard. For this purpose, a Keythely 2290E-5 5kV DC generator and a Keysight B2981A picoamperometer were used. After the averaging of the current measured at steady state conditions (i_{cond}), conductivity was obtained according to Eq. 1.

$$\sigma = \frac{J}{E} \ (S \cdot m^{-1}) \quad (1)$$

where J is the current density (given by the ratio between i_{cond} and the electrode surface S) and E is the electric field, set equal to 5 kV/mm.

III. RESULTS

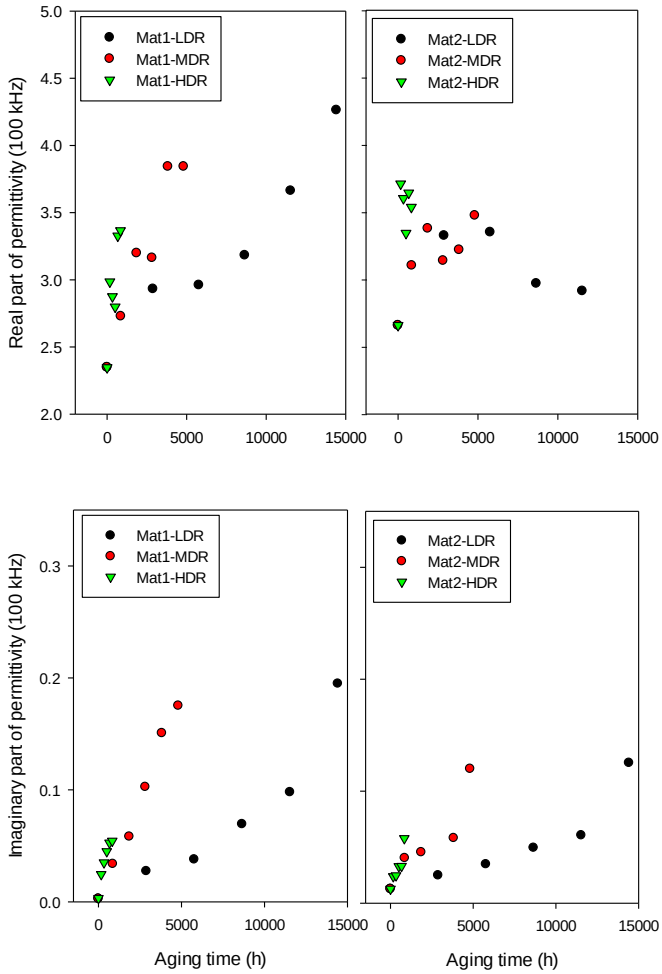

Figure 1 Real (a,b) and imaginary (c,d) parts of permittivity at 100 kHz for Material #1 (a,c) and Material #2 (b,d).

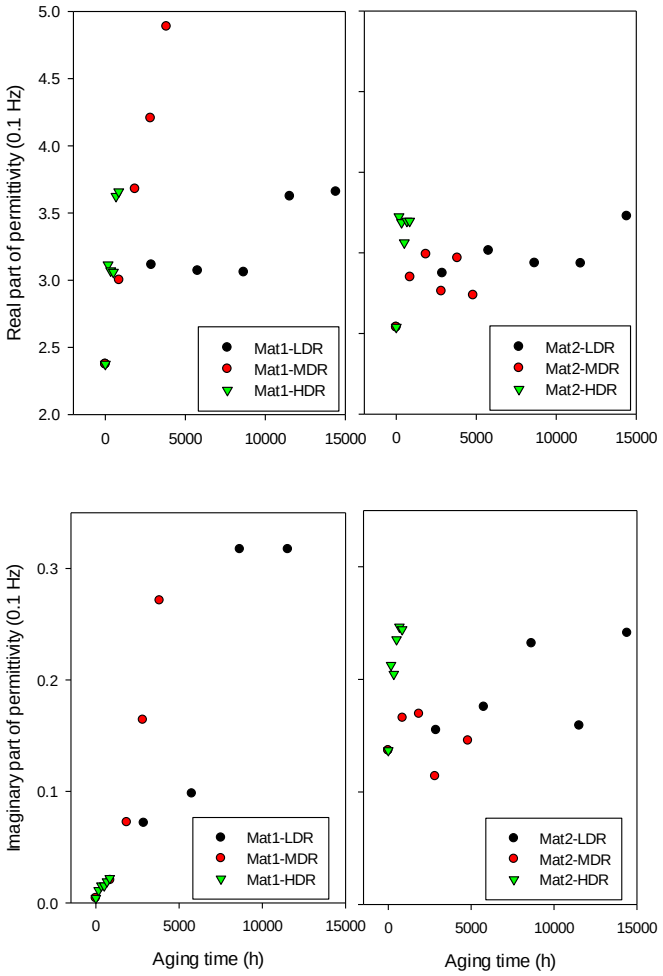

Figure 2 Real (a,b) and imaginary (c,d) parts of permittivity at 0.1 Hz for Material #1 (a,c) and Material #2 (b,d).

Complex permittivity values are reported in Figures 1-2 for the two investigated materials. Due to page constraints, it was chosen to report the values of both the parts of permittivity for two fixed frequencies: 100 kHz and 0.1 Hz. These frequencies were picked as representative of the highest and lowest frequency region of the measured dielectric spectrum, respectively. Moreover, due to the polarization mechanisms related to these frequencies, they can be considered as associated with the dipolar (100 kHz) and interfacial (0.1 Hz) polarization.

Material #1 complex permittivity (Figure 1.a, 1.c, 2.a, 2.c) is shown to increase with aging for both the analyzed frequencies. The increasing trend is shifted leftwards (towards lower aging times) as we increase the aging severity. Besides the variation of the property is bigger at lower frequencies (Figure 2.a, 2.c), one can notice that the raise is not monotonous with aging, e.g., depicting a step-like behavior of both parts of permittivity for the lowest dose rate.

The introduction of additives and fillers (Material #2) brings to a significant modification of the permittivity trends with aging [6-7]. At higher frequency (Figure 1.b, 1.d), the real part of permittivity does not follow a monotone increase with aging, independently on the aging severity. On the contrary, the

imaginary part exhibits a constant raise. As the aging severity increases, similarly to the previous case, the increasing trend is steeper and steeper. At low frequencies (Figure 2.b, 2.d), both the parts of complex permittivity do not show a monotone increase of the property, suggesting that this frequency is not easily relatable to the aging evolution of the filled material.

B. Conductivity results

Figure 3 reports the values of conductivity obtained for both the unfilled (Figure 3.a) and filled (Figure 3.b) XLPE material. From this figure, it is possible to notice a substantially different behavior between the two compounds. Focusing on unaged materials, one can notice that the presence of fillers and additives increase the conductivity value by 1.5 orders of magnitude. This was expected since additives, particularly ATH, are species which are known to influence also the conduction current [8]. With aging, conductivity increases up to 5 orders of magnitude in the case of non-filled polymer (Figure 3.a), while it increases by 2 orders of magnitude in the case of filled polymer (Figure 3.b). The cause of these different behaviors has to be found in the additive property of Mat#2, as it will be discussed in the following. With regards to the severity of the analyzed aging conditions, the different materials behave similarly to what already reported in the previous section.

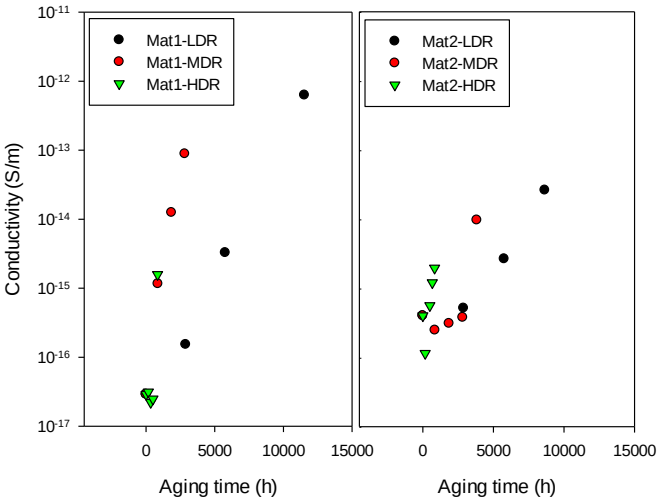

Figure 3 Conductivity on XLPE without additives (a) and XLPE with additives (b)

IV. Discussion

The using of different techniques, namely dielectric spectroscopy and conductivity allowed the definition of the electrical material response throughout a wide range of frequencies, down to DC. It has been presented that, depending on *f*, the property trends can be significantly different, particularly in the case of filled materials.

The analyzed frequency region in the dielectric spectra allows the definition of two different polarization mechanisms: dipolar and interfacial ones. The dipolar polarization, also called orientation polarization, is related to the movement of dipoles in the direction of the electric field. The frequency region, in which this polarization mechanism occurs, is estimated to be between 10^3-10^9 Hz, with a peak of dielectric losses at ~10^5 Hz. On the other side, the interfacial polarization, also called Maxwell-Wagner-Sillars (MWS) polarization, is linked to the accumulation of opposite charges at different surfaces (e.g., between the amorphous and crystalline phase or between matrix and additives) thanks to the external electric field. This kind of polarization, which is characterized by frequencies lower than ~10 Hz, is the most relevant in terms of dielectric losses, which can reach very significant values [6].

This clear division is done considering just a single relaxation phenomenon per each polarization mechanism. In real conditions, the material is made up of a copious number of molecules and interfaces, each one characterized by a specific relaxation time. This results into a convolution of different relaxation peaks in a single bigger peak from which is usually difficult to divide the different contributions. Nonetheless, each relaxation time τ is reported to be directly proportional to the size of the molecule and/or interface [9]. Hence, considering the frequency region analyzed in this work, we would be able to investigate the lower-frequency dipolar polarization (10^4-10^6 Hz) and the highest frequency interfacial polarization (10^2-10^{-2} Hz). This permits the investigation of big dipolar molecules and little interfaces, respectively. In particular, the former ones are the most important for aging investigation. As known, once subjected to aging stresses, macromolecular polymer chains initially face chain scission (Figure 4). This process creates two reactive smaller chains which can react with environmental oxygen creating a macromolecule with a permanent dipole (oxidized polymer chain). Indeed, while typical PE chains are characterized by no dipoles (estimated dipole moment ~0 D), the presence of bonded oxygen create a permanent dipole which can reach very high values of dipolar moment. As an example, a 12-carbon PE chain with a single oxygen atom i.e., ketone can exhibit ~3 D as estimated dipole moment. The created oxidized macromolecule responds electrically in the lowest dipolar polarization area due to, on the one hand, the presence of a non-zero dipolar momentum and, on the other hand, due to its big size. For this reason, it is evident how this frequency region well fits the aging evolution inside the polymer, as confirmed by the monotonous increase of the investigated properties for both materials (Figure 1).

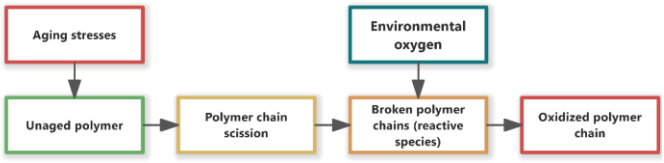

Figure 4 Schematic of oxidation reactions occurring in the polymer.

Nonetheless, the initial chain scission and the following oxidation process would modify the micro-interfaces inside the polymer insulation. Oxidation might lead to the modification of the local electrical properties (e.g., permittivity and conductivity) of the initial unoxidized polymer chain, due to the heighten electrical properties of the oxygen molecule. This local inhomogeneities could be the cause of the interfacial MWS polarization response already mentioned. Anyway, in this case, the created interfaces are very little in comparison to the one related to e.g., different materials. Because of that, the

aging-related interfaces are usually placed in the lowest frequency related to the interfacial polarization, as showed for Material #1 low-frequency response (Figure 2.a, 2.c). This is valid as long as the unaged material does not have intrinsically interfaces created by the presence of additives. Indeed, as the dielectric spectrum is made up of a convolution of different peaks i.e., different interfaces, the contribution of oxidative interfaces cannot be separated from the ones of other species e.g., additives. As an example, in the case here considered, the presence of antioxidants and, particularly, the huge concentration of ATH molecules brings to an abrupt increase of the low-frequency dielectric response of the unaged material by more than one order of magnitude (Figure 1.b). In particular, ATH molecules are characterized by an -OH bond which permit ATH molecules to arrange themselves into clusters by means of hydrogen bonds. This results into very big interfaces electrically responding in the lowest analyzed frequency region. As a result, the increase of the dielectric losses with aging, related to interfacial polarization is almost negligible and it does not regularly follow the polymer aging evolution (Figure 2.d). This suggests that the interfacial polarization response of the investigated polymer is completely lead by the additive interfaces, which may hide the new interfaces obtained from the aging process. On the other side, in the case of non-filled material, both the low and high frequency responses are very similar, and the complex permittivity trend perfectly follows the aging time.

Eventually, DC conductivity considers the effect of a copious number of charge carriers and impurities, in addition to the polymer degradation products created by aging. Undeniably, the aging effect is evident on conductivity, which raises as the aging time increases (Figure 3), due to the creation of oxidized species and double bond formation (e.g., C=C). This leads to e.g., a constant increase of the property for Material #1 (Figure 3.a) However, it may be difficult to single out the degradation contribution from these measurements, particularly if fillers are present in the compound. As an example, ATH fillers, as hydroscopic species, can easily bond with environmental moisture which is known to bring to a raise of the material conductivity [10]. This could be considered the reason for the constant value of conductivity for Material #2 (Figure 3.b) for aging times <500h.

All this being said, it is clear that, for unfilled polymers, the impact of the investigated frequency region is almost negligible, allowing the use of conductivity and broadband dielectric permittivity as aging markers. On the contrary, if the polymer owns a copious number of additives and fillers which can lead to e.g., bonded moisture and additional interfaces, the use of conductivity measurements and low-frequency dielectric response can be misleading. For this reason, the evaluation of the dielectric properties at higher frequencies, which are demonstrated to be mainly related to polymer dipolar species (e.g., oxidized polymer chain), is advisable.

V. Conclusions

In this article, analyses of some of the most common electrical techniques for aging evaluation are performed. In particular, the effect of the test frequencies is considered, permitting the choice of the most suitable one for the evaluation of the insulating aging evaluation. The results here presented highlighted the contribution of additives and contaminants on conductivity and dielectric spectrum, particularly in the low frequency region. This brought to an additional complexity in separating the contribution of the arising degradation-related species from the one related to additives. As a result, it has been concluded that, for non-filled materials, the trend of the chosen diagnostic quantity is the same throughout the analyzed frequency region. On the contrary, for filled materials, the dielectric response related to the dipolar polarization (highest analyzed frequency) is the only which ensure a monotone increase of the property with aging, since it has been showed to be associated with the degradation dipolar species (e.g., oxidized polymer chains).

Acknowledgment

The project leading to this application has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 755183. This publication reflects only the authors' view and the European Commission is not responsible for any use that may be made of the information it contains.

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