

Characterization of Ion Transport Properties in Synthetic Ester Oil by Polarization Current and Dielectric Spectroscopy

A. Cremasco^a, E. Logakis^b, M. Curti^a, and E. A. Lomonova^a,

^a *Eindhoven University of Technology, The Netherlands*

^b *ABB Corporate Research Center, Switzerland*

Abstract

The accurate estimation of the charge carrier mobility in synthetic ester oil-based insulation systems is required for the reliable prediction of the electric field induced by dc voltages. Some discordance is observed in the literature, therefore, this quantity is investigated by two alternative methods. First, the mobility is estimated in a material sample by measuring the polarization current and the ion time-of-flight during a voltage polarity reversal test. Next, the mobility is derived from the frequency-dependent loss tangent measured by dielectric spectroscopy. The values obtained by the two methods are consistent, in the range of $\approx 2\text{--}4 \times 10^{-10} \text{m}^2/\text{Vs}$ at 303 K.

1. Introduction

The transition towards renewable energies and the developments in high-power medium-voltage (MV) converters go hand-in-hand with the demand for compact MV components satisfying combined AC and DC insulation requirements [1]. In this respect, synthetic ester oil is an attractive choice, considering the needs of high dielectric strength, heat dissipation, operation safety, and low environmental impact [2].

The insulation design requires a careful analysis of the space charge distribution, to predict the electric field stress in the insulation. The charge transport model developed for HV-DC converter transformers insulated with mineral oil is used for synthetic ester oil insulated components [3, 4]. As shown in [5], an accurate estimation of the carrier mobility is required for the reliable prediction of the charge density accumulated at liquid-solid interfaces.

The ion mobility μ is difficult to quantify from macroscopic properties like the electrical conductivity, as the concentration of charge carriers in dielectric liquids is normally unknown. Several approaches have been developed to determine μ by applying an electric field across a material sample. These methods focus on the analysis of the dielectric response of the material in the time or in the frequency domain, by the polarization current test or by dielectric spectroscopy, respectively.

The literature on the mobility of charge carriers in MIDEI 7131 synthetic ester oil is not comprehensive as in the case of mineral oil [6, 7, 8]. The reference

values utilized in the literature span several orders of magnitude and are normally derived from current measurements at different electric field intensity: $\approx 10^{-7} \text{m}^2/\text{Vs}$ is found in [9] and utilized in [4, 10]; a value $\approx 10^{-10} \text{m}^2/\text{Vs}$ is found in another detailed work [11].

In this paper, the methods based on time and frequency domain analysis are briefly introduced in **Section 2**. The mobility at low electric field is estimated and compared with pre-existing results in **Section 3**, by the measurements of polarization current and dielectric spectroscopy, utilizing both parallel plate and cylindrical electrode arrangements.

2. Dielectric response analysis

2.1. Time domain

The mobility of charge carriers is estimated by analyzing the isothermal transient ionic current in the material, i.e., the polarization current [6]. The current is determined by firstly applying a step voltage to the test cell for a time interval t_0 sufficiently long, to sweep out the ions from the oil bulk and collect them at the electrode surfaces. The voltage polarity is then reversed: a current peak is observed in the polarization current after a time interval τ_m from the polarity reversal. This interval is considered approximately equal to the ion time-of-flight across the gap.

The mobility μ is found assuming the liquid as a symmetric weak electrolyte exposed to an electric field $E < 0.1 \text{ kV/mm}$, so the effect of the space charge is reasonably neglected [12]. Therefore,

$$\mu = \frac{L^2}{V\tau_m} k_E, \quad (1)$$

where L is the gap length and V is the amplitude of the voltage step. The coefficient k_E is a shape factor, which depends on the electrode arrangement: $k_E = 1$ for plate electrodes, whereas for concentric electrodes of inner electrode radius r it holds

$$k_E = \left(\frac{1}{2} + \frac{r}{L} \right) \ln \left(1 + \frac{L}{r} \right). \quad (2)$$

If $r/L > 2.4$, the shape factor can be reasonably neglected as $|k_E - 1| < 0.01$.

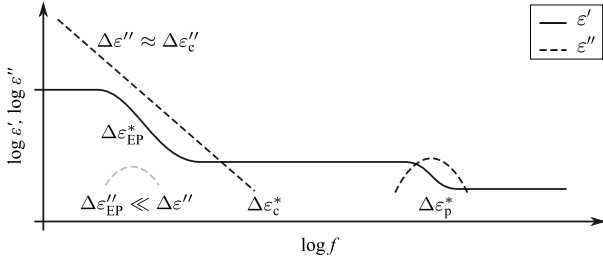


Fig. 1 – (a) Dielectric response spectrum revealing polarization ($\Delta\epsilon_{EP}^*$, $\Delta\epsilon_p^*$) and conduction ($\Delta\epsilon_c^*$) processes.

The main advantage of this method is the simple relationship between μ and the measured quantities. However, several preliminary tests are required to define a suitable interval t_0 and make the results independent from it [6].

2.2. Frequency domain

The mobility of charge carriers is estimated by analyzing the dielectric response of the material in the frequency domain. The response is represented by the complex permittivity $\epsilon^* = \epsilon' - j\epsilon''$, which is defined as $\mathbf{D} = \epsilon^* \epsilon_0 \mathbf{E}$, where ϵ_0 is the dielectric permittivity of vacuum, $\mathbf{E}$ is the electric field and $\mathbf{D}$ is the dielectric displacement; a typical spectrum is shown in **Fig. 1**. The analysis focuses on the low-frequency range (< 10 Hz), where two main processes occur: charge conduction and electrode polarization. The conduction due to mobile charge carriers is revealed as a constant unit negative slope in the spectrum of ϵ'' plotted in a log-log graph, as shown by $\Delta\epsilon_c^*$ in **Fig. 1**. Polarization processes appear both as an increase in ϵ' and a peak in ϵ'' , as shown by $\Delta\epsilon_p^*$. In this respect, a peak due to electrode polarization is rarely visible at low frequencies, as it is masked by the dissipation of the conduction process, as shown by $\Delta\epsilon_{EP}^*$.

The mobility of ions is estimated by analyzing the dynamics of electrode polarization, caused by the growth in the concentration of mobile carriers at the electrode of opposite polarity. This process is evident at low frequencies, for which ions have enough time to migrate and accumulate. As a result, the dissipation factor $\tan \delta = \epsilon''/\epsilon'$ exhibits a knee at the highest frequency allowing an appreciable charge accumulation at the electrodes [6, 8]. Similarly to (1), the frequency f_c is related to V , L and μ as

$$\mu = L^2 \frac{f_c}{2} \frac{\pi}{2V} \approx 0.8 \frac{L^2 f_c}{V}. \quad (3)$$

The factor $f_c/2$ is included to consider the ion transit time $\approx 1/2f_c$, whereas $2V/\pi$ is the average voltage applied over the semi-period.

3. Measurements

3.1. Sample and electrodes

The synthetic ester oil used in this analysis is MIDEI 7131. The samples were degassed at 353 K, 5 mbar

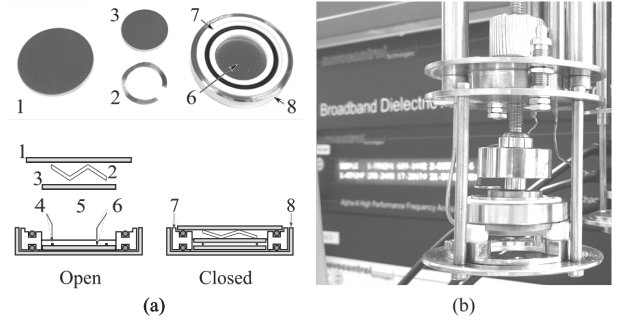


Fig. 2 – (a) BDS-1308 parallel plate sample cross-section: cell closing plate (1), spring (2), upper electrode (3), silica spacer (4), sample liquid (5), lower electrode (6), teflon insulation and seal rings (7), cell carrier plate (8); (b) BDS 1308 parallel plate sample in the test cell.

for 96 h and stored in a borosilicate glass jar.

Two measurement electrodes designed by *Novocontrol Technologies* are considered in this work:

- BDS-1307 cylindrical electrodes with guard rings: $L = 2$ mm, $r = 8$ mm ($k_E = 1.004$) [13];
- BDS-1308 parallel plate electrodes with 20 mm diameter and $L = 100 \mu\text{m}$, adjusted by silica spacers ($k_E = 1$) [14].

The test cell, which is shown in **Fig. 2**, is positioned during the measurements in a cryostat. The temperature T is stabilized at 303 K by the *Novocontrol* Quatro Cryosystem and using dry nitrogen, i.e., $\Delta T < 0.2$ K and $dT/dt < 0.2$ K/min.

3.2. Polarization Current

The measurement is performed by the *Keithley* 6517 Electrometer, which is utilized also as the voltage source. The sample is pre-polarized at $-V$ for $t_0 = 2$ h, which is found to be the minimum time required to make τ_m independent from the pre-polarization time t_0 in both cells. The choice of V is dictated by the maximum acceptable E during the test. Besides, V must be sufficiently high to limit τ_m , to ensure enough ions reaching the opposite electrode and originate a current peak before recombining.

Measurements for the cylindrical and the parallel plate electrodes are presented with the same scale in **Fig. 3**. As shown in **Fig. 3(a)**, the current peak in the cylindrical test cell at $E = 0.025$ kV/mm is the least pronounced due to the longer τ_m , which allows more carriers to recombine. The amount of recombined ions is reduced to enhance the current peak by raising $V = 250$ V and decreasing τ_m , as shown in **Fig. 3(b)**; as a drawback, $E = 0.125$ kV/mm, which exceeds the limit of the weak field assumption ($E < 0.1$ kV/mm [12]). The current measured with the parallel plate cell is shown in **Fig. 3(c)**, the peak is well visible even at low E thanks to the shorter L (0.1 mm) and τ_m , since fewer ions recombine while crossing the gap.

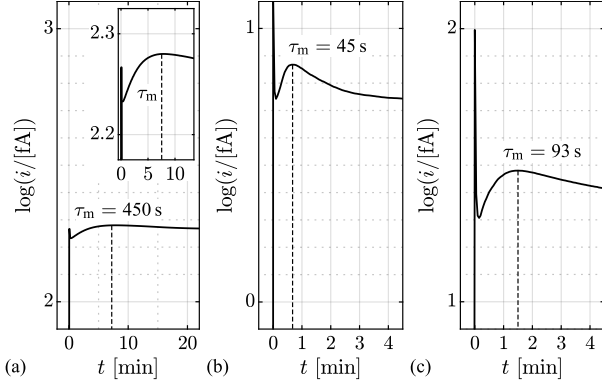


Fig. 3 – Measured current after polarity reversal: BDS-1307 cylindrical cell with (a) $V = 50$ V and (b) $V = 250$ V, and (c) BDS-1308 parallel plate cell with $V = 1$ V.

The values of μ determined by both test cells according to (1) are reported in **Table 1**, together with results obtained in previous works [9, 11]. The measurement performed at $V = 250$ V in the cylindrical cell (0.125 kV/mm) exhibits a slightly higher μ compared to the measurements in the parallel plate cell (0.01 kV/mm), possibly due to the increased E . This difference is acceptable for practical purposes, but highlights the challenge of using $L \geq 2$ mm with synthetic esters due to $\mu \approx 10^{-10} \text{ m}^2/\text{Vs}$.

Results from [9] are obtained by a needle-plate electrode configuration at relatively high E compared to the other cases. Values of μ extrapolated from the $\sqrt{I} - V$ characteristics assuming space charge limited conduction are two orders of magnitude higher than the values obtained in [11] and in the present work. This difference can be attributed to the onset of electric field enhanced dissociation of ion pairs and electro-hydro dynamic turbulence that increases the apparent mobility of ions [12, 15]. The results presented in this work confirm those obtained at low E in [11], where a cylindrical electrode configuration is adopted.

3.3. Complex Permittivity

The measurement of ϵ^* is performed by the *Novocontrol* Alpha-A High Performance Frequency Analyzer and the BDS-1308 parallel plate test sample. The real and imaginary part of ϵ^* are plotted in **Fig. 4**.

Table 1 – Mobility estimated by polarization current test. Electrode type: N = Needle, P = Plate, C = Cylindrical.

Test cell	L [mm]	V [V]	$E \times 10^6$ [V/m]	τ_m [s]	$\mu \times 10^{10}$ [m ² /Vs]
(C) BDS-1307	2	50	0.025	450	1.8
		250	0.125	45	3.6
(P) BDS-1308	0.1	1	0.01	93	1.1
(N-P) [9]	5	$> 10^4$	> 2	N/D	$> 10^2$
(C) [11]	1	40	0.04	223	1.12

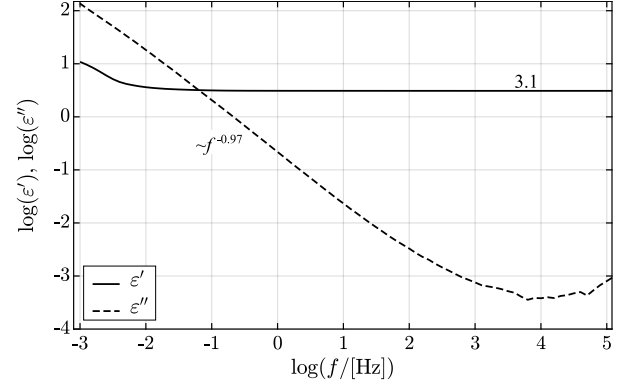


Fig. 4 – Frequency spectrum of ϵ' and ϵ'' measured by dielectric spectroscopy with the BDS-1308 parallel plate cell.

The characteristic is qualitatively similar to the one shown in **Fig. 1**: the onset of a relaxation mechanism at the higher frequencies is revealed by a positive slope in ϵ'' from $f = 100$ kHz, and is normally caused by the orientation polarization of the larger molecular dipoles in the liquid. At lower frequencies, the dissipating effects are dominated by charge conduction, as indicated by a plateau in ϵ' and the constant negative slope in ϵ'' . The onset of electrode polarization is visible in the spectrum of ϵ' at $f \approx 10$ mHz. The contribution of electrode polarization to the total dissipation in the dielectric is negligible compared to charge conduction, so no visible changes are present in the slope of ϵ'' .

The estimation of μ is performed analyzing $\tan \delta$, which is shown in **Fig. 5(a)**: the plot presents a negative unit slope in the frequency range where charge conduction dominates, before the onset of electrode polarization; a similar behavior is observed for mineral oils in [6]. The plot is magnified in correspondence with f_c in **Fig. 5(b)**; the values of μ estimated by (3) are indicated in **Table 2**. Values of μ obtained by the dielectric spectroscopy are similar to the ones extracted from the polarization current and τ_m .

The main advantage offered by dielectric spec-

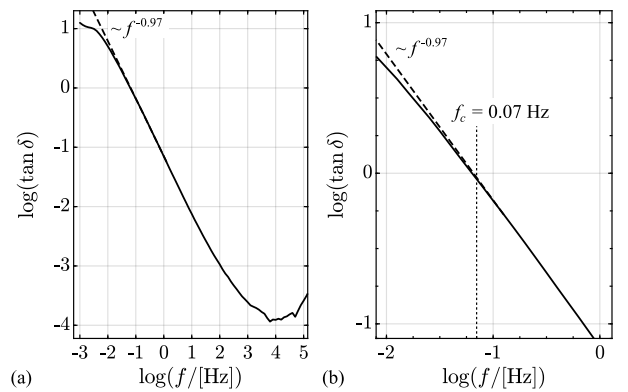


Fig. 5 – (a) $\tan \delta$ with dashed line indicating negative unit slope and (b), $\tan \delta$ magnified in correspondence with f_c .

Table 2 – Mobility estimated by dielectric spectroscopy.

Test cell	L [mm]	V [V]	$E \times 10^6$ [V/m]	f_c [Hz]	$\mu \times 10^{10}$ [m ² /Vs]
BDS-1308	0.1	1.4	0.014	0.07	4.0

troscopy is that the sample does not need to be pre-polarized, and t_0 does not need to be determined. A measurement down to 0.01 Hz with ten points per decade is sufficient for this material and the BDS-1308 test cell ($f_c = 0.07$ Hz), it takes ≈ 3 h. However, the interpretation of results is not as straightforward as the analysis of the polarization current peak.

4. Conclusion

The values of charge carrier mobility μ in MIDEI 7131 synthetic ester oil available from the literature range from 10^{-7} to 10^{-10} m²/Vs at ambient temperature. Such a large range limits the confidence in the results predicted by models relying on this property and possible design criteria. The mobility at 303 K is estimated in this work by different test cells and methods. First, the polarization current after polarity reversal is analyzed to measure the ion time-of-flight; the test is performed considering both a parallel plate and a cylindrical electrode sample cell. The values of μ extracted by this method are in the range of 1.1 – 3.6×10^{-10} m²/Vs. Next, the dielectric response of the oil is analyzed by dielectric spectroscopy. The equation relating μ with the onset frequency of electrode polarization is proposed, a value $\mu = 4 \times 10^{-10}$ m²/Vs is found, confirming the applicability of dielectric spectroscopy for this investigation. These results provide a further experimental reference for μ in MIDEI 7131 synthetic ester, enabling a more accurate modeling of charge transport in this material.

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6. References

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