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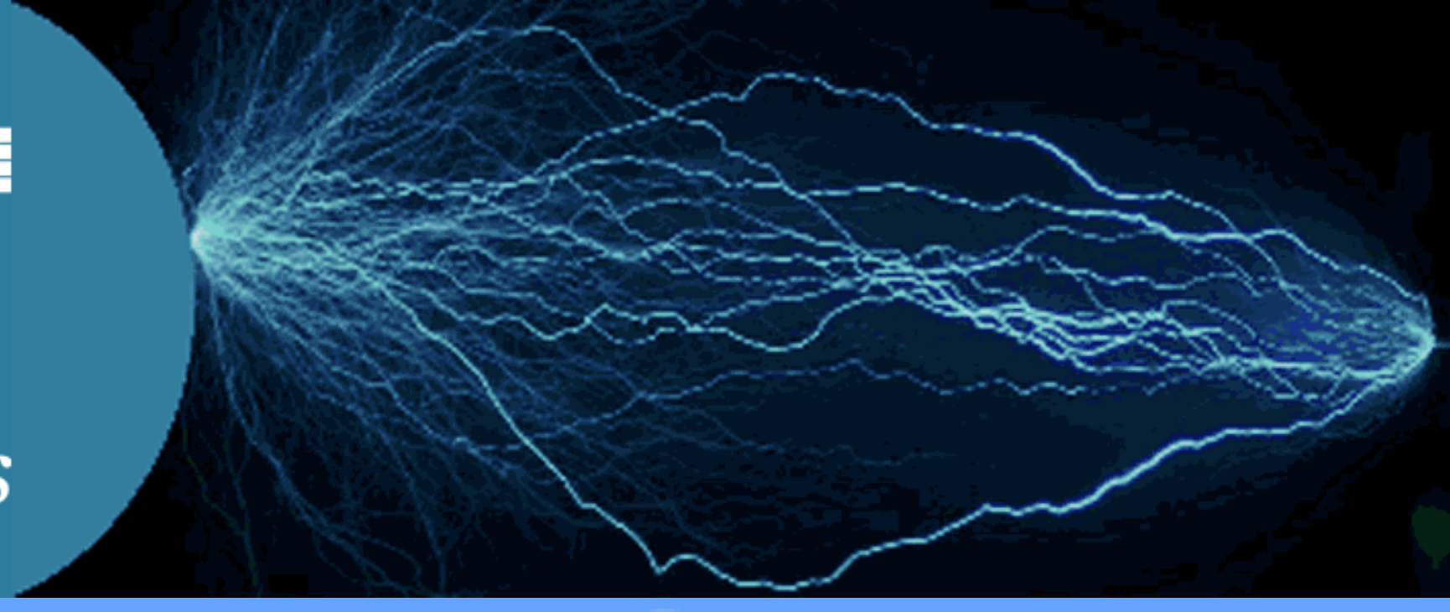

# Absorption Current Behaviour of Polyethylene/Silica Nanocomposites

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## Introduction

- Absorption current is considered to be one of the important characteristics of polymers with regard to their time-domain response to a DC poling field.
- The presence of nanofiller/polymer interfaces in nanocomposites will affect the current flow due to the introduction or modification of the charge trapping sites; charge carriers may move easily via nanoparticle interfaces, depending on their characteristics.
- This paper reports on an investigation into the absorption current behaviour of PE nanocomposites containing 0 wt%, 2 wt%, 5 wt% and 10 wt% of $SiO_2$ nanofiller, either untreated or treated using trimethoxy(propyl)silane.

## Experimental

- The base polymer: PE composed of 80 wt% of LDPE and 20 wt% of HDPE.
- The filler: nano-$SiO_2$, either untreated or treated using trimethoxy(propyl)silane.
- Nanocomposites were prepared using a solution blending method.
- The test samples were melt pressed at 150 ˚C, followed by isothermal crystallization at 115 ˚C.
- The thickness of each sample was 200 µm.
- Absorption current measurements were performed using a Keithley 6487 Picoammeter / voltage source and a sample holder with 20 mm diameter electrode.
- A step voltage of 40 $kV_{(DC)}$ $mm^{-1}$ or 25 $kV_{(DC)}$ $mm^{-1}$ was applied.
- Prior to measurement, each sample was grounded at 60 ˚C in vacuum for at least 72 h so that excess charge would dissipate.

## Absorption Current Behaviour at 40 kV $mm^{-1}$ DC Field

- Figure 1 shows the plot of time dependence absorption current for all samples at a constant DC field of 40 kV $mm^{-1}$ over 1 h at room temperature.
- All the nanocomposites showed a current-time characteristic that is different from unfilled PE.
- The slope data at the beginning of the test and at times around 200 s onwards are summarized in Table 1, calculated assuming the power law relationship:

$$I = At^{-b}$$

*I* = current
*t* = time after the application or removal of the external voltage
*A* = temperature dependent factor
*b* = constant representing the slope of the log-log current-time plot

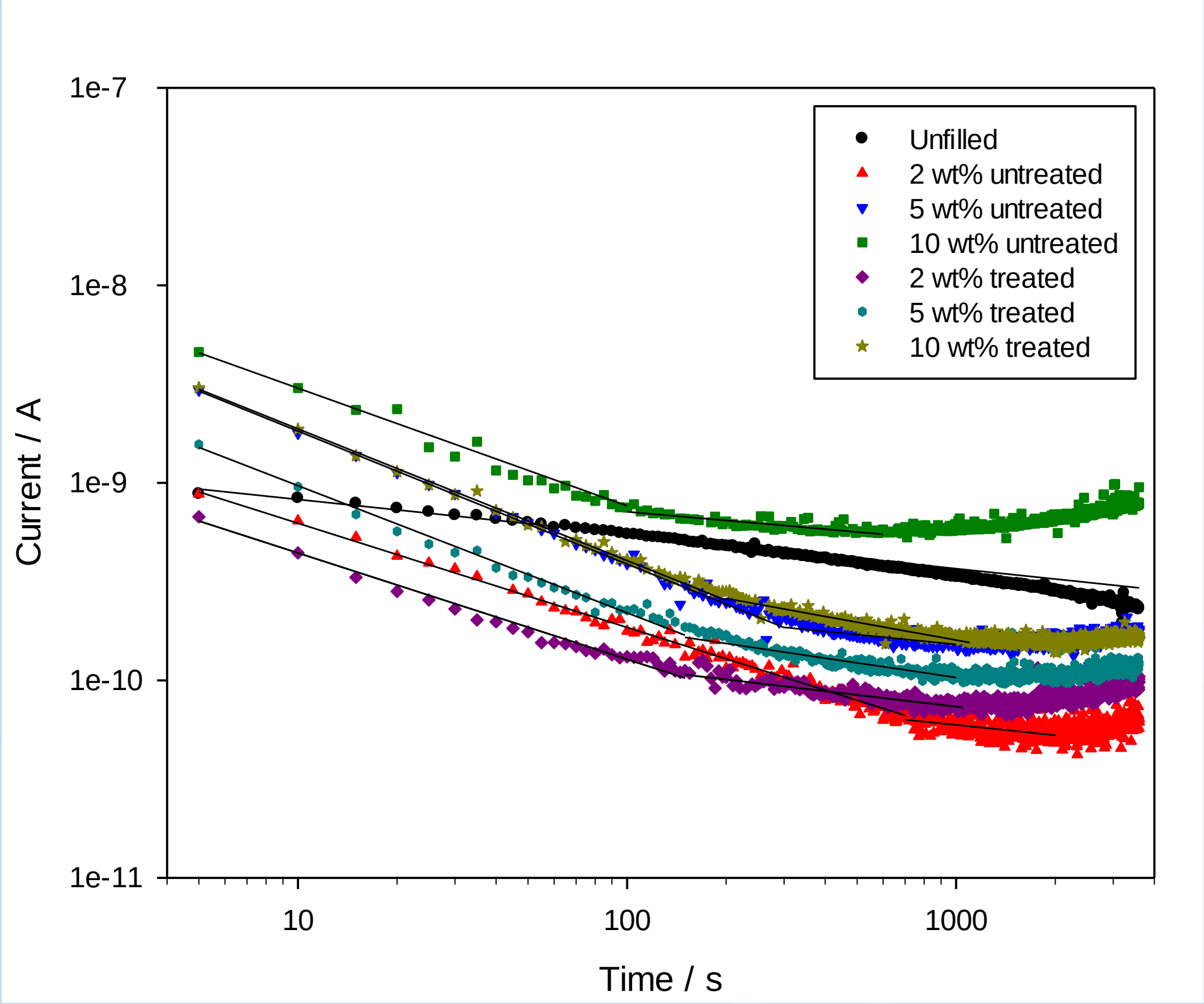

**Figure 1:** Plot of absorption current against time up to 1 h for all investigated samples at an applied field of 40 kV $mm^{-1}$. Repeated experimental runs showed that data variation within a factor of 3 is typical. Two lines are fitted for nanocomposites to indicate a change in slope prior to a current rise.

- The slope of all nanocomposite data was steeper at the beginning of the test compared with the unfilled PE.
  - This indicates that the incorporation of nano-$SiO_2$ may have increased the mobility in the nanocomposites, thus leading to faster decay of the current as the carriers take shorter time to reach the electrodes.

**Table 1:** Slope calculated from the absorption current data at the beginning of the test and around 200 s onwards at an applied field of 40 kV $mm^{-1}$.

| Sample | Slope, *b* | |
|---|---|---|
| | 0 s – 200 s | 200 s onwards |
| Unfilled | 0.18 | 0.18<br>(unchanged) |
| 2 wt% untreated | 0.53 | 0.17 |
| 5 wt% untreated | 0.67 | 0.16 |
| 10 wt% untreated | 0.60 | 0.15 |
| 2 wt% treated | 0.54 | 0.20 |
| 5 wt% treated | 0.64 | 0.25 |
| 10 wt% treated | 0.67 | 0.30 |

- It is noteworthy that 0 ≤ *b* ≤ 2 is consistent with dipole orientation, carrier tunnelling and carrier hopping while 0 ≤ *b* ≤ 1 is consistent with charge injection forming trapped space charge.
- While nanoparticles with their large surface areas may act as additional electron traps, conversely, they may decrease the average hopping distance relative to that of the polymer matrix and thus increase the mobility.
- All the nanocomposites exhibit a characteristic reduction in slope at times around 200 s onwards; the gradient of the plot then becomes comparable to that exhibited by the unfilled PE.
  - This could indicate a change in dominant mechanism of the absorption current.
- The change of slope could also be interpreted in this way: electronic transport (electron transfer) is effectively controlled by trapping, i.e. an electron may travel rapidly through the system for a short time, but its effective or average mobility is greatly reduced as a result of being immobilized for much longer period in localized states (traps).
- The curious rise of the current was observed for all nanocomposites near the end of the test.
  - This could be due to interfacial polarization caused by the nanofiller/polymer interfaces, not being compensated by the charge behaviour.

## Charge Carrier Mobility

- The mobility of charge carrier is highly dependent upon the release of charges from deep traps in addition to the underlying current caused by a large number of shallower traps introduced by the nanoparticles.
- All the nanocomposites have higher charge carrier mobility than the unfilled PE (see Figure 2), calculated using the formulae:

$$\mu = \frac{0.786d^2}{t_p V}$$

*μ* = mobility of charge carriers
*d* = sample thickness
$t_p$ = the time at which absorption current is at its maximum or the time at which a slope change occurs
*V* = applied voltage

- This could be associated with the presence of shallower traps that are related to the nanofiller/polymer interfaces in addition to the original trap distribution that characterizes the crystalline/amorphous interfaces. The former serve to assist in charge transport.

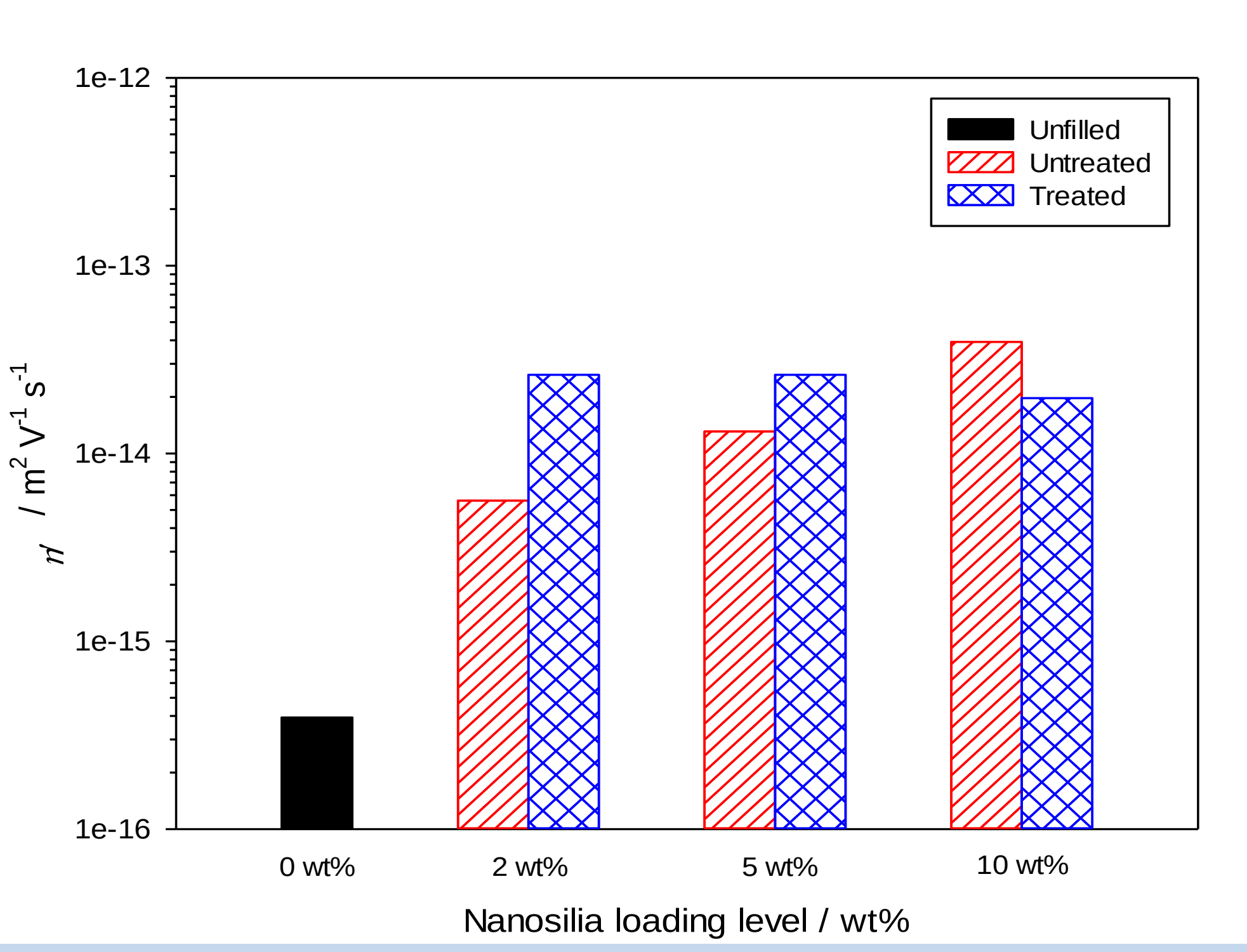

**Figure 2:** Charge mobility of unfilled polyethylene and nanocomposites containing different types and amounts of nanosilica

## Absorption Current Behaviour at Different DC Fields

- Figure 3 compares the current-time characteristics of the nanocomposite samples obtained at a longer duration at two different DC fields, i.e. 40 kV $mm^{-1}$ and 25 kV $mm^{-1}$.

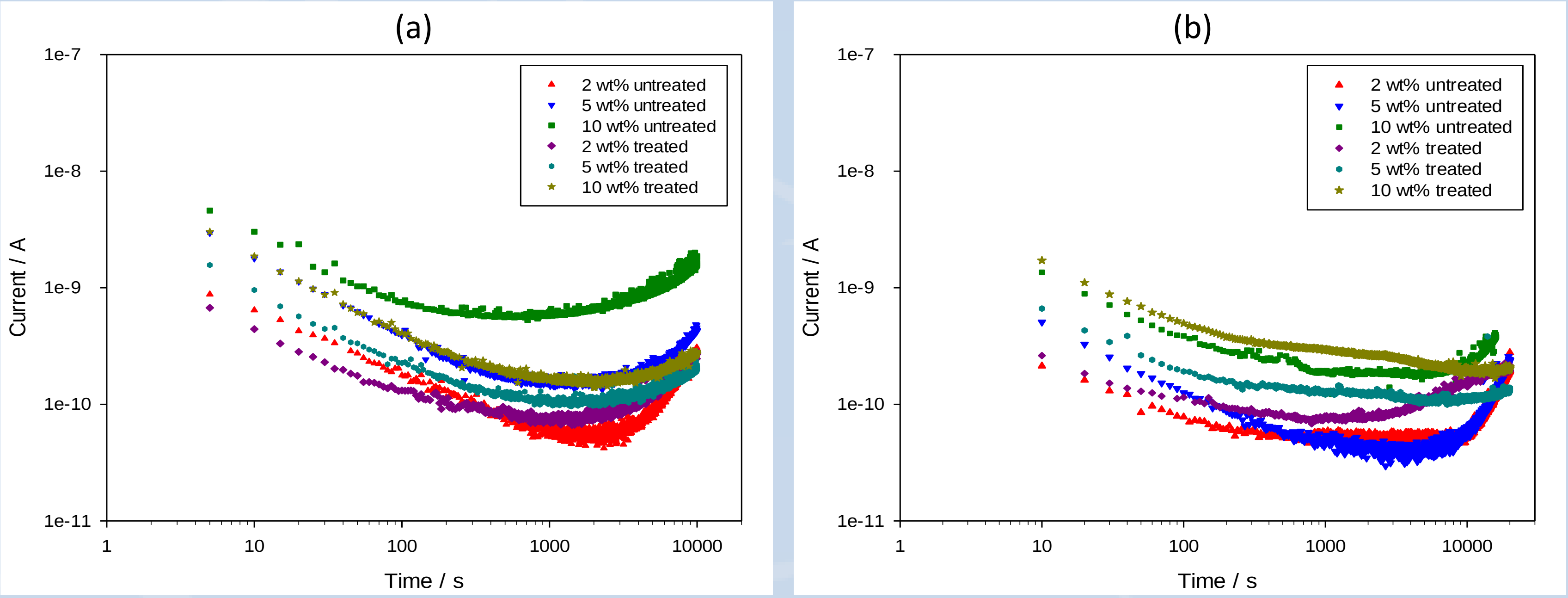

**Figure 3**: Plot of absorption current against time for all nanocomposites at an applied field of (a) 40 kV mm-1 up to $(1 \times 10^4)$ s, (b) 25 kV mm-1 up to $(2 \times 10^4)$ s

- Interfacial polarization mechanism in the nanocomposites seems to have happened at later times at reduced DC field.
  - The increase of current is generally not noticeable in nanocomposites at a poling time of $10^4$ s at an applied field of 25 kV $mm^{-1}$, but was noticed at poling times of $10^4$ s to $10^5$ s.
  - The absorption current characteristics at 25 kV $mm^{-1}$ applied field is different for the case of nanocomposites containing treated nano-$SiO_2$ and nanocomposites containing untreated nano-$SiO_2$ at higher filler loading (5 wt% and 10 wt%).
  - The increase of current happened earlier in the nanocomposites containing untreated nano-$SiO_2$ than the nanocomposites containing treated nano-$SiO_2$.
  - This indicates that the interfacial polarization mechanism in the nanocomposites containing treated nano-$SiO_2$ is different from that in the untreated counterparts at reduced DC field.

## Conclusions

- While the current behaviour through the unfilled PE decreases with time in a conventional manner, all nanocomposites reveal an initial decrease followed by a period in which the current increases with increasing time of DC field application.
- The inclusion of nano-$SiO_2$ into PE causes the slope of the absorption current to be steeper than that of the unfilled PE at the beginning of the test, highlighting the possibility of increased charge mobility in the nanocomposites.
- At a reduced DC field, the nanocomposites containing treated nano-$SiO_2$ exhibited different behaviour from the untreated counterparts.
- Presently, literature into such current-time characteristics of absorption current data is scarce and more work is necessary fully to explain the underlying charge transport mechanisms in nanocomposites.

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