SPATIAL DEPENDENCE OF ELECTRIC FIELDS DUE TO SPACE CHARGES IN FILMS OF ORGANIC DIELECTRICS USED FOR INSULATION OF POWER CABLES

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Abstract

We measured the electric fields due to space charge within film samples of several commercial polyolefins used as medium voltage power cable insulation. We charged the films at 60°C with DC voltages applied for several hours. For dry charged films, vacuum deposited gold or aluminum provided the electrodes. For wet charged films, one or both surfaces of the film were bare and in contact with an aqueous 0.05 molar solution of NaCl. We measured the fields using the thermal pulse (TP) method. To use this method with the wet charged films, we painted colloidal graphite electrodes onto the films after they were charged. The results show that thermoplastic polyethylene (PE) charges negatively for both wet and dry charging. Crosslinked polyethylene (XLLPE) charges negatively when dry charged, but small amounts of either positive or negative charge were found after wet charging. Water tree retardant crosslinked polyethylene (TRXLPE) charged positively for both dry and wet charging. A crosslinked (ethylene-propylene-diene) terpolymer (EPDM) formulation charges negatively when dry charged and positively when wet charged. The EPDM formulation was pyroelectric. This pyroelectricity was absent when the clay filler was removed from the formulation.

Introduction

Recent work [1,2] has shown that the thermal pulse method of measuring space charge fields is a powerful tool for the study of the electrical properties of films of polymeric insulation materials. In that work, we showed that charge distributions in polyethylene and ionomers (sodium or zinc salt neutralized copolymers of PE and a metallocene acid) consistently became nearly uniform for steady state, constant charging current conditions. Also the charge distributions were shown [2] to be consistent with a simple model [3] of homopolar charge injection and transport, or with extensions of that model to include bipolar injection and transport [4].

In this paper, we extend the work in references 1 and 2 to include other materials used as solid insulation in medium and high voltage power cables. We also explore the effects of wet charging with aqueous NaCl solutions. These data provide insight into the charging processes in dielectric polymers. Also, TP measurements may be able to characterize the chemical compositions of polymers in ways that relate to their service performance.

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Experimental

The films used here were compression molded at Union Carbide Corporation from commercially available materials. Films for dry charging were outgassed for several hours in an evaporator. Electrodes were vacuum-deposited on both surfaces through a mask. Gold worked well for the crosslinked films but distorted the thermoplastic PE. In such cases aluminum was used because it could be deposited at lower temperatures. The films were mounted and held taut between closely fitted concentric brass rings. The ring-mounted films were charged in a thermally insulated metal can in contact with a temperature regulated hot plate. The bottom electrode rested on a grounded magnetic support and the top electrode was contacted by a thin metal ribbon drawn to the support by a small magnet. The sample temperature was measured with a thermocouple in contact with the grounded support. We used as high a voltage as we felt the films could withstand up to a maximum of 3500 V. The charging current typically decreased for several hours until it reached a constant steady state value. We then turned off the voltage and quickly placed the film on a heat sink to quench it to room temperature.

Films for wet charging were mounted as received between standard 9 mm glass joints held together by a spring clamp. Room temperature vulcanized silicone rubber formed the seal between the glass joints and the film. The tubular glass extensions of the glass joints were bent at 90° to make a U shaped assembly when the joints were attached to the film. Each half of the assembly was filled with a 0.05 molar NaCl aqueous solution. Copper disks soldered to the ends of separate wires and immersed in the salt solutions provided electrical contact between the surfaces of the film and the voltage supply. The U shaped glass assembly was placed in an oven for charging. The conduction in the films was greater in the salt solution than when dry. This necessitated lower voltages for wet than for dry charging. Films were removed from the holder, dried with a paper towel and mounted in rings like those used for the dry charged films. Aqueous colloidal graphite electrodes were painted on each surface of the film and allowed to dry in ambient air. These colloidal graphite electrodes were each 10-25 μm thick and had a resistance of a few hundred ohms. Table 1 lists characteristics and charging conditions for representative samples.

We used the TP method to obtain transients containing information about the amount and distribution of space charge in the films. This method is described in a series of papers [1,2,5,6]. In this method a nominally 1 μs heat pulse is applied to the electrode on one side of a few mil thick polymer film. Non-uniform thermal expansion of the film during thermal equilibration moves different parts of the embedded space charge in predictable ways, producing a transient flow of charge between surface electrodes on the film. We measure this charge vs time under short circuit conditions. A battery is used to calibrate [1].

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TABLE I. Description of Samples and Charging Conditions

<table>
<thead>
<tr>
<th>Code</th>
<th>Material</th>
<th>d (µm)</th>
<th>C (pF)</th>
<th>Tc (°C)</th>
<th>ts (hrs)</th>
<th>Ec (MV/m)</th>
<th>Jc (pA/cm²)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>131</td>
<td>PE</td>
<td>70</td>
<td>54</td>
<td>60</td>
<td>15.5</td>
<td>8.0</td>
<td>--</td>
<td>Wet, total 47µm graphite electrodes painted on for measurement.</td>
</tr>
<tr>
<td>144</td>
<td>PE</td>
<td>45</td>
<td>66</td>
<td>60</td>
<td>22.5</td>
<td>22.0</td>
<td>0.69</td>
<td>Dry, aluminum electrodes.</td>
</tr>
<tr>
<td>139F</td>
<td>XLPE</td>
<td>170</td>
<td>20</td>
<td>60</td>
<td>92</td>
<td>8.8</td>
<td>7.2</td>
<td>Dry with graphite painted on gold to increase heat.</td>
</tr>
<tr>
<td>19</td>
<td>PE</td>
<td>50</td>
<td>59</td>
<td>57</td>
<td>19</td>
<td>50.0</td>
<td>4620</td>
<td>Dry, gold electrodes, noisy charging current.</td>
</tr>
<tr>
<td>118</td>
<td>TRXLPE</td>
<td>62</td>
<td>80</td>
<td>60</td>
<td>24</td>
<td>24</td>
<td>--</td>
<td>Wet, gold side (+), bare side (-).</td>
</tr>
<tr>
<td>117</td>
<td>EPDM formulation</td>
<td>108</td>
<td>30</td>
<td>60</td>
<td>24</td>
<td>6.5</td>
<td>--</td>
<td>Wet, bare side (+), gold side (-).</td>
</tr>
<tr>
<td>137</td>
<td>EPDM formulation</td>
<td>155</td>
<td>30</td>
<td>60</td>
<td>20</td>
<td>16.1</td>
<td>--</td>
<td>Dry, gold electrodes.</td>
</tr>
<tr>
<td>134</td>
<td>EPDM formulation less Clay</td>
<td>53</td>
<td>58</td>
<td>60</td>
<td>5</td>
<td>6.7</td>
<td>--</td>
<td>Wet, bare elec., not pyroelectric.</td>
</tr>
<tr>
<td>142</td>
<td>EPDM formulation less Clay</td>
<td>76</td>
<td>48</td>
<td>60</td>
<td>4</td>
<td>13.1</td>
<td>3.5</td>
<td>Dry, gold elec., not pyroelectric.</td>
</tr>
</tbody>
</table>

The transient amplitude in units of potential difference across the film, V0, or equivalently in units of the associated charge on the electrodes, Q0 = CV0, where C is the measured capacitance of the film. The initial values of the calibrated transients for both surfaces give directly the net space charge and the first moment of the distribution [1]. A more careful analysis [5] using a fit of the transients to a series of exponentials gives several Fourier coefficients of the electric field distribution and its derivative, the space charge distribution. For this paper we use the simplified direct analysis of the transients to describe the nature of the charge distribution near the electrodes and the predominant sign of the net space charge.

**Results and Discussion**

**PE**

Transients for dry charged PE (sample 144) are shown in Figure 1. This material acquired a net negative charge. The electric field from this space charge is stronger at the - than at the + electrode, in accord with our previous results [2]. This result implies that electron injection dominates the charging process. A larger field at the injecting than at the ejection electrode is consistent with the simple charge injection model [3,4]. Rounding of the transient at short times shows some net + charge at the + electrode, which indicates hole injection. A monotonically decaying transient for the - electrode shows that there is no net + charge there. The total space charge field is about 38% of the charging field for 144.

Wet charged PE (sample 131, not shown) also acquired a net - charge with a larger field at the - than at the + electrode. The space charge field was a larger fraction of the charging field for wet charged films indicating that the salt water enhanced electron injection. Details of the charges near the electrodes are obscured by the thermal mass of the graphite electrodes.

**XLPE**

Dry charged XLPE (sample 139F) also acquired a net + charge, as shown by the transients in Figure 1. However, the space charge field in this case is much stronger at the + electrode (25% of the charging field) than at the - electrode (5% of the charging field). This is partly due to a large amount of + charge at the - electrode, indicated by rounding of the transient at short times. This + charge at the injecting (-) electrode should enhance - charge injection and give greater - charge density at the + electrode than in PE. Positive charge at the - electrode is probably not due to holes. Rather, we speculate that + ions are responsible for this charge in XLPE. We have not modeled mobile ions sufficiently to predict results for this case.

Data for two wet charged XLPE samples (not shown) were inconsistent and we cannot draw conclusions for this case. We suspect the inconsistencies were due to a faulty power supply, rather than something inherent in these samples.
The transients for the dry charged TRXLPE (sample 19 shown in Figure 3) show a net + space charge throughout the film. This is just opposite the result for the polyethylenes. The net space charge field is about 13% of the charging field and due to + space charge. Note that the transient for the + electrode in Figure 3 goes positive at short times showing that - space charge is concentrated at the + electrode. This result is difficult to interpret by a simple charge injection model since the charge adjacent to both electrodes is heterocharge. However a reasonable conclusion is that hole injection dominates the conduction process, giving the net + space charge, and that ions contribute to the heterocharge at both electrodes. The absence of the blocking effect of homocharge at the injecting + electrode would greatly enhance injection in this case, and could account for the fact that the charging currents are orders of magnitude larger for the TRXLPE than for the polyethylenes.

Wet charged TRXLPE (sample 118, not shown) has the same general charging characteristics as the dry charged film, but with less homocharge at the + electrode, and a more uniform distribution of + charge. The space charge field was at least 10% of the charging field for this case.

**EPDM Formulation**

This formulation includes EPDM, red lead, zinc oxide and a surface activated clay filler. Like PE, the dry charged formulation (sample 137 shown in Figure 4) acquired a net - charge with some + homocharge at the
The space charge field at the - electrode is about 9% and 15% of the charging field at the + and - electrodes respectively. Additionally, this EPDM formulation has a pyroelectric response giving an offset between the baseline and the transients at long time. This response is most probably due to permanent polarization of some component in the formulation. The field from this polarization is a few % of the charging field.

The wet charged EPDM formulation (sample 117, not shown) behaves quite differently from dry charged formulation, giving a net + space charge, and some negative homocharge at the - electrode. While this is the first clear example of significant differences between wet and dry charging, we can not rule out the possibility that the differences in outgassing of the films may be partly responsible. It is reasonable to expect differences in charge injection behavior between the cases of charging with aluminum electrodes and charging with salt water electrodes.

EPDM Formulation Without Surface Activated Clay

To identify the source of the permanent polarization that we found in EPDM, we measured several dry charged formulations 1) without red lead, 2) without zinc oxide, and 3) without surface activated clay filler. These gave very similar transients for the injecting (-) electrode with somewhat greater variability for the + electrode. This consistency supports the idea that the space charge behavior is controlled primarily by the nature of the polymer, and not by the additives. The permanent polarization is present in all these formulations except for the one without clay. The dry charged formulation less clay (sample 142, shown in Figure 5) showed no pyroelectricity and no positive charge at the + electrode as was found in the original formulation. The space charge fields were 14% and 60% of the charging fields at the + and - electrodes respectively. This was the most highly charged of the series of films measured here.

The wet charged EPDM based formulation from which the clay had been left out (sample 134, shown in Figure 6) had a net + charge. This is the reverse of the results for this material when dry charged, and consistent with the results for the original EPDM formulation.

Calculated Distributions

The above discussions were based on the shapes of the TP transients. These transients can be analysed by curve fitting procedures [5,6]. As examples, the PE 144 transients were fit using the first six Fourier coefficients and the calculated electric field distribution is shown in Figure 7. This distribution is typical of the polyethylenes we have measured and has the general shape predicted by a simple model of negative charge injection and transport [3]. As another example, XLPE 139F was fit using coefficients 1,2,4,6 and 7 and yielded the field and charge distributions shown in Figures 8 and 9. Note there is fairly uniformly distributed negative charge except for the heterocharge at the - electrode, mentioned above. This more formal analysis gives a better indication of the magnitude and spatial extent of this charge than do the transients themselves. Some of the oscillations in the calculated distribution are artifacts of the fitting procedure. Also the fitting procedure dictates that the calculated space charge be zero at the electrodes. The simple model predicts that the injected charge is maximum at the injecting electrodes.
Figure 8. Electric field distribution for dry charged XLPE calculated from the transients in Figure 2 using Fourier coefficients 1, 2, 4, 6 and 7.

Figure 9. Space charge distribution for dry charged XLPE calculated from Figure 8.

Conclusions
The TP method gives a sensitive and reliable way of characterizing the charging behavior of dielectric polymers. Polymers of different chemical composition give different characteristic TP transients when DC charged to steady state. The results are repeatable from sample to sample of the same material. Differences are seen in charging behavior between metal and salt water electrodes. The method also gives valuable insight into the physical mechanism underlying the conduction and charging phenomena in these materials.

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Commercial products are referred to in this paper to adequately describe the work that was done. Such referral in no way implies endorsement by the National Institute of Standards and Technology (formerly NBS).

References