Thermoplastic Materials Aging under Various Stresses

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***Abstract*— The most popular cable insulation material used is XLPE due to its excellent electrical and thermal properties. However, it does not lend itself to ease of recycling. As a result of an increase in concern worldwide regarding environmental protection, it is the objective of this work to investigate whether a thermoplastic material could be used to replace XLPE for cable insulation. Among thermoplastic materials, HDPE is regarded as one with the most similar properties as XLPE. Although it is clear that the performance of polymeric material changes with different stresses, especially polymer nanocomposites aging process under AC electric field stresses, there are also not many publications on how a superimposed AC voltage would affect the insulation's performance in HVDC power systems.**

This paper reports the dielectric properties of HDPE under thermo-electrical stresses. DC stress with and without a superimposed AC stress were applied in the experiments undertaken. The degradation of materials with change in frequencies are summarized and discussed.

Keywords— dielectric spectroscopy, electrical properties, FTIR-ATR, HDPE, thermo-electrical aging

I. INTRODUCTION

Although cross-linked polyethylene (XLPE) is the most popular cable insulation material with its excellent electrical and physical properties [1], it may not be a good choice of cable insulation in the future as it is not easy to recycle. Therefore, it is necessary to find suitable alternatives for XLPE [2]. In general, thermoplastic materials are good candidates.

With the continued development of power transmission systems and the increase in voltage level for high voltage direct current (HVDC) power cables, it raises a new challenge for insulation materials to be easily recycled when they are used in underground cables and subsea cables.

HVDC power transmission systems invariably have power electronics embedded. Voltage Source Converters (VSC) are the technology of choice due to its lower volume size and simpler structure [3] and also its capability in power factor compensation [4]. A disadvantage of VSC is the resulting waveform will be a DC voltage combined with transients generated from the switching in the kHz frequency [5]. The combined voltage may cause further degradation. Hence, it would be useful to investigate the effect of such composite voltage waveforms on the insulation material.

Many researchers investigated the performance of oil-paper insulation under DC, AC, impulse individually or combined. The partial discharge characteristics of gas cavity in oil-pressboard insulation under AC and DC combined stress was reported in [6-7], which showed that, as DC component increases in the AC & DC combined stresses, the amount of PD charge would decrease and partial discharge inception voltage would increase.

Some publications demonstrated the electrical breakdown properties of oil-paper insulation under AC-DC combined voltages. Yan Wang et al. [8] showed the breakdown characteristics of oil-impregnated paper under AC&DC combined stress. The results showed that oil-paper insulation demonstrated high DC breakdown strength but low AC breakdown strength. The value of AC&DC breakdown strength was between DC and AC. Yan Wang et al. [9] evaluated breakdown performance of the oil-paper aged AC and DC combined voltage stresses. The research kept AC voltage, DC voltage and AC superimposed with DC voltage as a constant. The results showed that as DC component increased in the combined voltage stresses, breakdown strength of oil-paper insulation increased.

However, there are not many publications on polymers aged under AC and DC combined stresses, and this is the motivation for the current investigation. To evaluate the performance of polymer insulation aged under AC and DC combined stresses, thermo-electrical aging studies were carried out on HDPE samples. The DC voltage 6 kV was applied across the sample with a superimposed sinusoidal AC voltage of various frequencies. In this paper, the superimposed frequency was 1 kHz, 2 kHz and 3 kHz. Fourier Transform Infrared Spectroscopy - Attenuated Total Reflection (FTIR-ATR) measurements and dielectric spectroscopy will be carried out before and after aging.

II. EXPERIMENT SET-UP

High-density polyethylene (HDPE) is a common thermoplastic material, whose properties are similar to XLPE as both of them have high molecular weight, compared with other PE materials.

The melting temperature T_m is 128 °C, through Differential Scanning Calorimetry (DSC) measurement. The result is shown in Fig.1.

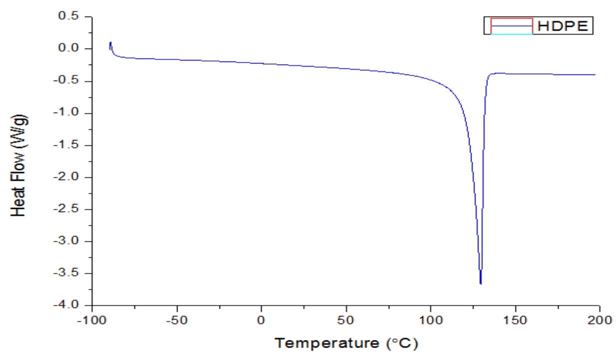

Fig. 1. DSC result of HDPE

The HDPE film, 50×50 mm square and 50 um in thickness, was sandwiched between the electrodes, as shown in Fig.2.

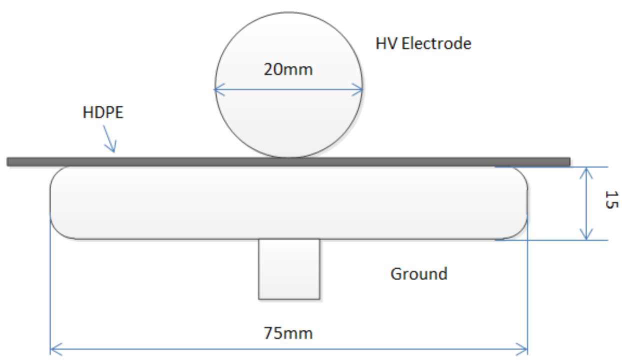

Fig. 2. Electrode of thermo-electrical aging

Both electrodes are made of stainless steel. The 20 mm diameter sphere electrode was connected to the high voltage power supply, while the lower electrode was connected to ground.

To simulate the actual condition present in a cable insulation system, thermal stress was applied to the samples as well. Therefore, the electrodes and sample arrangement were placed in an oven so that the surrounding temperature was controlled.

To model the long-time degradation of cable insulation in a short time, it is necessary to do accelerated aging. To accelerate aging, the temperature should be above the XLPE cable operation temperature 90°C [10]. However, the aging temperature should be below the melting temperature (128°C) of HDPE. Therefore, the temperature was set at 95°C.

AC voltage and DC voltage was applied across the sample simultaneously. The combined waveform is shown in Fig.3.

The 6 kV DC voltage was generated by a power amplifier (model 30/20A-H-CE produced by TRec Inc.). The AC voltage was generated by a signal generator (Tektronix model AFG 3102C) and input to the power amplifier set at 3000 amplification. The frequencies of the 3 kV AC peak voltages applied were 1 kHz, 2 kHz and 3 kHz.

To describe the combined voltage stress, the definition of ripple factor is [9]:

$$AC\% = \frac{V_{AC}}{V_{DC}} \times 100\% \tag{1}$$

where, V_{DC} is the mean value of DC voltage, V_{AC} is the peak value of AC voltage.

In this paper, ripple factor is 50%.

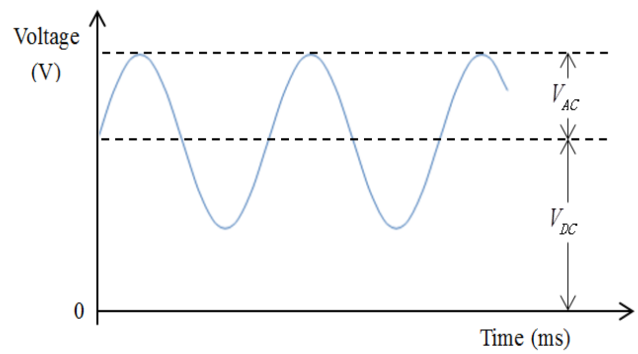

Fig. 3. AC&DC combined voltage waveform

Dielectric spectroscopy measurement and FTIR-ATR measurement were used to evaluate the un-aged and aged HDPE samples. The broadband dielectric spectroscopy, model Concept82 (and produced by NOVOCONTROL company) has gold-plated electrodes and a frequency range of 3 uHz to 3 GHz.

An FTIR-ATR, (model Nicolet iS 16 produced by Thermos Fisher Company), was used to measure the absorption by the samples of wavenumbers between 4000 cm^{-1} and 350 cm^{-1}.

III. Results and Discussions

A. FTIR-ATR Measurement Results

The FTIR-ATR results of un-aged and aged HDPE are shown in Fig.4-6.

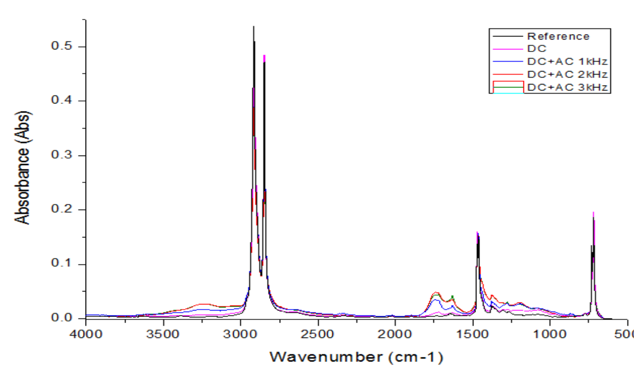

Fig. 4. FTIR-ATR results of HDPE

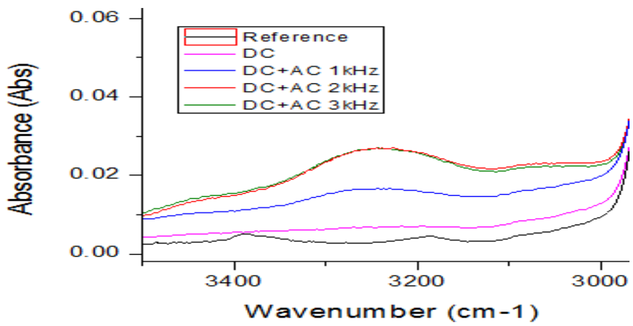

Fig. 5. Zoomed-in FTIR-ATR spectrom zoomed-in (OH-group)

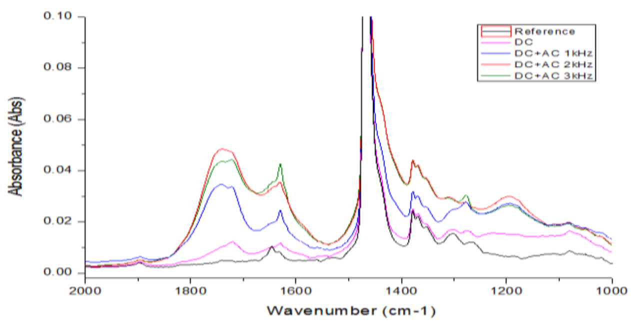

Fig. 6. Zoomed-in FTIR-ATR spectrum zoomed-in (carbonyl group and CO group)

The original HDPE sample consisted of methylene groups, which can be described by four peaks at 2918 cm^{-1}, 2850 cm^{-1}, 1462 cm^{-1} and 718 cm^{-1}, respectively [11].

Compared with the original HDPE sample, new peaks at 3226 cm^{-1} (see Fig.5), 1727 cm^{-1}, 1629 cm^{-1} and 1206 cm^{-1} (see Fig.6) emerge on the curves of aged samples. Among them, 3226 cm^{-1} peak can describe the O-H group. According to the complexity of the shape of the OH region, it probably contains both acidic OH and non-acidic OH as well as "water". Peak at 1727 cm^{-1} is carbonyl groups (C=O). As there is not a single peak at 1727 cm^{-1}, it probably contains carbonyl of 2 types - one that is part of the COOH acid and other likely non-acidic ketone type. The 1206 cm^{-1} peak is C-O.

The intensity of absorbance can demonstrate the amount of molecular groups. Sample aged by DC stress only is not very different from the original sample. It is obvious that the absorbance of samples aged by AC combined with DC stress at 3226 cm^{-1}, 1727 cm^{-1}, 1629 cm^{-1} and 1206 cm^{-1} are much higher than the original sample. In addition, with frequencies increase, the absorbance increases. This means there are more O-H group and carbonyl groups generated under AC combined with DC stress compared with DC stress. When AC voltage and DC voltage were combined to age the sample simultaneously, a higher frequency was probably responsible for the generation of new groups.

The new molecular groups were probably created as a result of decomposition of the original HDPE. Under thermal stress and electrical stress, chain scission of the long chains in HDPE may result. Oxidation may take place at the new terminals, thus creating O-H groups and C=O groups. The more the new groups were created means the more the molecular chains were broken and the worse the degradation became.

Therefore, combined stresses can age a sample further, and the degradation becomes worse with an increase in frequency.

B. Dielectric Spectroscopy Measurement Results

The dielectric results of HDPE are shown in Fig.7-9 with real permittivity, imaginary permittivity and dissipation factor respectively.

Real permittivity is regarded as the dielectric constant of materials. For original HDPE and aged HDPE, ε' is a constant between 2.30 and 2.55 for frequencies between 10^{-2} to 10^{-4} range. In each curve, ε' slightly increases as the frequency decreases. This result agrees with the expected variation of the dielectric constant with frequency. The orientation polarization of the polar molecule is influenced by frequency and to a certain extent, the dielectric constant decreases as frequency increases [5].

The value of ε' of the sample aged under DC voltage stress is a little lower than the un-aged sample but not too much different, while AC&DC combined stresses aged HDPE are much higher than the un-aged HDPE. When aged by AC&DC combined stresses, and with frequency increases, the constant increased.

When polymer is aged, cross-linking and degradation will happen simultaneously [12]. The performance will highly depend on both of the procedures. Cross-linking will establish strong links between molecules which make molecules closer and hard to orient under external electrical stress, which will lead to orientation polarization to be low. As the orientation polarization is low, the dielectric constant will be low as well. Along with degradation mechanism, the original long molecule chain will break, polar molecules may be created, such as water and low molecular acid, which will increase the orientation polarization and increase the dielectric constant further.

The aging mechanism depends on AC voltage frequency. It is obvious from Fig.7 that DC voltage stress encourages cross-linking and therefore the dielectric constant is lower than the original HDPE. Since AC and DC combined stresses encourage the degradation process, it therefore follows that the dielectric constant is higher than the original HDPE. Higher AC frequency will encourage further degradation. When samples are aged under higher aging AC frequency, more polar molecules will be created, which will result in the dielectric constant to increase further. The results are consistent with the FTIR-ATR results.

As shown in Fig.9, tan δ values of aged samples are higher than the un-aged HDPE in the frequency range from 10^{-2} to 10^{4} Hz. tan δ remains a constant for frequencies between 10^{0} to 10^{4} Hz and dramatically increases with frequency deceases in the frequency range from 10^{0} to 10^{-2} Hz. The tan δ of sample aged by AC&DC combined stress is higher than the sample aged by DC stress only. In addition, with increases in the aging AC frequency, the value of tan δ increases.

In the high frequency range, as time cycle is short, there is not enough time to establish relaxation polarization. Therefore, in high frequency range, electron polarization is the main polarization type and there is no relaxation loss. While in the low frequency range, as the applied electric field of dielectric spectroscopy is not shifted so quickly, the time cycle is enough for relaxation polarization to establish.

The dielectric results are in accordance with the FTIR-ATR results.

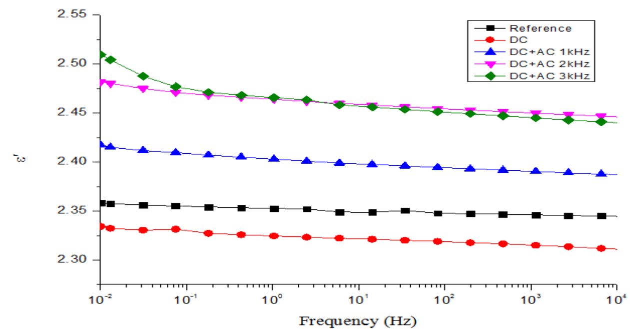

Fig. 7. Real permittivity of HDPE

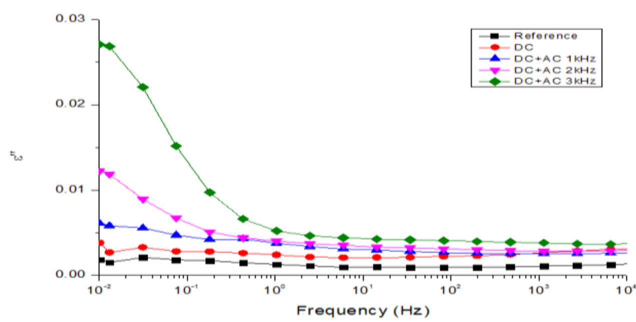

Fig. 8. Imaginary permittivity of HDPE

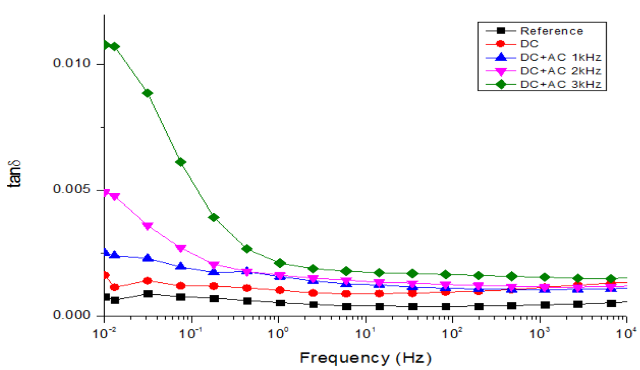

Fig. 9. Dissipation factor of HDPE

IV. CONCLUSIONS

When HDPE materials are aged under AC&DC combined stresses with various aging frequencies, the results of dielectric spectroscopy and FTIR-ATR can be concluded as: DC stress may lead to cross-linking. Less OH group and carbonyl group created under DC stress compared with DC&AC stress. AC stress may lead to polymer degradation, which will lead to more OH and carbonyl group created. As AC frequency increases, polymer will be degraded further, which means, more OH group and carbonyl group will be created and the dielectric constant and dissipation factor will increase.

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