

Article

Modelling Electroluminescence Emission in Polymeric Material Using Dimensional Analysis Method

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Abstract: The Electroluminescence (EL) technique has garnered significant interest over time for its reliable outcomes that enrich our understanding of the onset of electrical degradation in polymeric material. There is growing interest in combining both the EL technique and space charge measurement methods to obtain a more profound knowledge of the degradation and ageing of insulation. The generation of charge carriers in polymeric materials is widely acknowledged to be a crucial aspect of EL, involving injection, de-trapping, and field dissociation processes. This research investigates the variables that influence how EL emissions behave, including applied voltage or electric field, applied electrical frequency, ageing of material, and selection of materials and gases employed. It was found that these factors can be helpful in predicting the process of electrical ageing in insulation to avoid any catastrophes that may occur. A mathematical approach relating these factors and the intensity of EL is proposed through the aid of Dimensional Analysis method. A close relationship is obtained that suggests this mathematical approach can be utilised as a tool to predict electrical ageing of insulation material.

Keywords: electroluminescence; ageing; insulation; modelling; dimensional analysis



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1. Introduction

A dielectric material is an insulation material that prevents damage to electrical devices from excessive voltages. It can be in the form of solids, liquids and gases. The qualities necessary in an excellent insulating material include high resistivity, low electrical conductivity, strong dielectric and thermal strength, and low dielectric loss. Most commonly used insulators are polymeric materials and some incorporate additives such as antioxidants, stabilisers, plasticisers, and cross-linkers in the raw materials to produce better materials. Any polymer, however, will eventually degrade when exposed to repeated electrical and physical stresses.

It has been accepted [1–3] that high voltage cables typically experience electrical degradation at the surface or at the bulk of the insulating material. It is believed that this phenomenon mainly occurs because the manufacturing of the material and the construction of the high voltage cable is imperfect, contributing to possible defects within the cable such

as poor adhesion, voids, inclusion and screen protrusion. However, the presence of these defects in insulating material can now be avoided with increasing awareness, control and cleanliness in the process of manufacturing high voltage systems.

The presence of space charges in polymeric insulating material has also been identified as the main factor that leads to the deterioration and ageing of the material. Holes, electrons, charged particles, and ions are all examples of space charges. Under a strong electrical field, space charges can be transferred into the insulating material and trapped within the material. Over the time, the space charges accumulation can enhance the polymer's electric field, especially at the metal-polymer interface, leading to the polymer's electrical degradation, which usually exhibits a tree-like growth known as treeing.

Under the influence of high voltage stresses, water treeing may occur in polymer in the presence of water. However, water treeing does not directly lead to the electrical breakdown of polymer but acts as a starting point for generating electrical treeing that causes a total breakdown in the polymer. Electrical treeing can generate from water treeing due to temperature, voltage transient, chemical changes inside a water tree, electric stress, and type and length of the water tree [4]. Electrical treeing can also grow in a dry environment at high voltage stresses. Electrical treeing growth is continuous and cannot be stopped, which eventually leads to the breakdown of polymer.

This undesirable problem has encouraged numerous efforts [5–7] mostly using non-destructive methods to predict the time-to-failure of the material. Among these methods are the commonly used Pulsed Electro Acoustic (PEA), Thermal Pulse Method (TPM), Laser Induced Pressure Pulse (LIPP), and so forth. The main purpose of these methods is to measure the distribution of space charge in the material to predict or detect the early occurrence of electrical treeing within the insulating material of the polymer before a total breakdown can occur.

Until recent years, a new but promising technique for detecting electrical ageing, particularly at the interfacial region has been implemented. This technique, called the electroluminescence (EL), observes the existence of light emission in the insulating material. Electroluminescence occurs due to the excitation of atoms in the material when high electrical voltage is applied to it. It is stated that the changes in the excitation states are an early indicator of electrical ageing in dielectric material [8]. The visible and ultraviolet regions of the EL spectrum correspond to the regions where EL occurs in the material due to enhanced electrical stress [9].

This paper looks at variables that influence how EL emissions behave; including applied voltage or electrical field, applied electrical frequency, the ageing of the material, the types of polymers and surrounding gases employed. This paper is presented as follows; Section 1 provides an introduction on EL, Section 2 outlines the mechanism of EL, and Section 3 explains the method of detecting EL emission. The factors that influence the EL intensity are identified in detail in Section 4 followed by the EL technique and other space charge measurement methods in Section 5. Sections 6 and 7 then discuss the proposed EL modelling and the simulated results before concluding the paper in Section 8.

2. Mechanism of Electroluminescence

The light emission phenomenon or electroluminescence (EL) can be caused by the interaction of mobile and trapped charges in the polymeric material upon the application of an external electric field [2]. EL occurs in most polymers under DC, AC, and impulse stress. There have been some arguments as to whether EL is the cause or the consequence of electrical treeing. However, many have demonstrated that EL occurs before the emergence of electrical trees and, in some circumstances, before partial discharges [1–3,9–11]. The sequential excitation and relaxation of the polymer molecules' valence electrons are closely related to the EL generation process.

2.1. Excitation Process

In general, there are two forms of EL excitation; the direct high-field EL and the hot electron excitation. In direct high-field EL, the external electrical field directly caused the excitation of the electronic state. This process includes the Zener effect, in which electrons travel from the valence to the conduction band of the material or, in certain circumstances, it also enables electrons to jump across the ground-state impurity level to the conduction band. This technique, however, is not relevant to large-band gap materials such as polyethylene, since the electrical field necessary to elicit this behaviour is substantially greater than the polyethylene breakdown field [2].

The impact mode or hot electron excitation is the second form of excitation. As explained intensively by Laurent et al. [2], the applied electric field will provide sufficiently large kinetic energy to accelerate injected or de-trapped charge carriers in the material. Inelastic collisions will occur between carriers and molecules of material that cause them to be excited. The interaction between electrons and holes with the centre of the luminescence can potentially absorb a considerable amount of energy from the electrical field, thus generating ionized molecules. When charge carriers collide with each other, an electron is produced and it will gain enough energy to accelerate by application of an electric field and cause more collisions, leading to an electron avalanche [2]. However, this technique is only practical when the polymer has a considerable free volume and highly energized injected electrons.

Another mode of excitation that has been observed by many researchers recently [1–3,12] is known as bipolar charge recombination. This excitation can occur at a low electrical field where trapped space charge can be excited electronically by recombination with charge carriers of opposite polarity. Under DC stress application, charge carriers can be infused into the polymer from both electrodes. These space charges will migrate across the polymer and interact with charge carriers of opposite polarity and hence recombination of charge carriers can occur. Under AC voltage, injected holes cannot de-trap during the positive half-cycle and remain in the interface region. These trapped holes and injected electrons recombine in the negative half-cycle to generate an excited electron-hole pair [2].

2.2. Relaxation Process

For EL to be produced, de-excitation or relaxation of charge carriers must occur after the excitation process takes place. The relaxation process can be in the form of a chemical or physical reaction [2]. In the chemical reaction, excited states release their stored energy in the form of fragments of molecules or free radicals, both of which can undergo subsequent chemical events. In a physical reaction, no further chemical reaction is produced as the excited centre falls back to its ground state. Emissions of fluorescence or phosphorescence might result from this completely reversible phenomenon [2].

3. Detection of EL

Currently, there are two methods of detection of EL emission [2]; diverging field and uniform field configuration. In diverging field configuration, the needle electrodes are embedded in a bulk polymer. This configuration can locally produce a constant field and can escalate until the breakdown of the polymer if the electrical stability of the specimen is maintained. Meanwhile, the uniform field configuration utilizes plane parallel electrodes and thin polymer films in the experimental setting. Many kinds of research [8,13–21] opted for this configuration because it can be used for many types of samples, as the former configuration only applies to polymers that can be made with embedded electrodes [2].

Both configurations are reported to deliver different results. In a uniform field configuration, EL emission occurs at low fields; 2 to 20 kV/mm [22,23], and in diverging field configuration, EL emission is detected at much higher fields; >200 kV/mm [24,25]. Owing to the configuration of the plane parallel electrode, the injected electrons and trapped holes recombine at the surface of the polymer which resulted in the electroluminescence emission

at this interface area instead of in the bulk of the polymer. These processes lead to charge migration into the polymer bulk where EL emission can be observed at much higher fields.

4. Factors Affecting the EL Intensity

Dodd et al. [18] suggested that the electroluminescence emission during the initial phase of AC electrical breakdown can be a good experimental tool to charge injection processes, interface failure, electron avalanche phenomenon, and the partial discharge activity.

There are numerous factors that may affect the intensity of light emissions such as the amount of voltage applied, sample ageing, types of electrodes, distance of needle tip to the electrode, additives and aromatic group sample, time of voltage application, presence of oxygen in sample, frequency applied, types of the voltage waveform and many more. We will discuss some of the factors in this section and their effect on polymeric materials such as low density polyethylene (LDPE), crosslinked polyethylene (XLPE), polyethylene terephthalate (PET), polypropylene (PP), polytetrafluoroethylene (PTFE) and polyethylene naphthalate (PEN).

4.1. EL vs. Applied Voltage

It has been accepted by many researchers [1,8,26,27] that the amount of voltage or electrical field supplied to a polymer, either in impulse, DC, or AC stress, influences the electroluminescence intensity. When the supplied voltage increases, it increases the number of charge carriers injected at the bulk of the polymer. This eventually increases the potential of holes recombining with electrons in every half-cycle, resulting in greater EL intensity. As previously stated, free radicals are formed when charge carriers and polymer molecules collide in the polymer. The amount of the free radicals may increase with the increasing voltage applied, and as long as the local field is greater than the critical field for charge injection, this can initiate electrical treeing [1].

Bamji et al. [26] undergo EL observation on XLPE by increasing the applied voltage up to 28 kV and decreasing it back to 16 kV varying from 30 Hz to 600 Hz. In each fixed operating frequency, it is identified that as the applied voltage increases, the intensity of EL emission increases as well but with a different threshold voltage onset of electroluminescence as illustrated in Figure 1. The EL intensity also decreases when the applied voltage is decreased but stops when the voltage drops below the light inception level. It is claimed that EL emission is constant and only ceases when the voltage is withdrawn or lower than the threshold level [26]. The influence of applied voltage on EL emission is similar for any experimental conditions, whether EL is observed at or above room temperature, in liquid-impregnated material or under various gaseous environments. EL in polymer displays a threshold-like behaviour where it rises linearly as applied voltage is increased above its inception voltage. However, the EL counts and the EL inception voltage may vary for each condition.

The EL threshold of PEN is observed to be much lower at approximately 30 kV/mm, with a gradual increase in EL with field [28]. An electroluminescence experiment was also carried out on BOPP and XLPE at room temperature with samples subjected to 50 Hz AC stress at a high voltage field until 223.4 kV/mm for BOPP and 57 kV/mm for XLPE. Although the EL intensity rises with increasing voltage, it showed a decreasing trend at each level under constant voltage, with a steeper decline at higher fields [29]. The same phenomenon was observed for Bisphenol A epoxy resin, PP and polystyrene (PS) when subjected to 50 Hz AC and DC applied voltages under a divergent field. The EL intensity showed a gradual increase with increasing voltage until the initiation of the electrical tree [30].

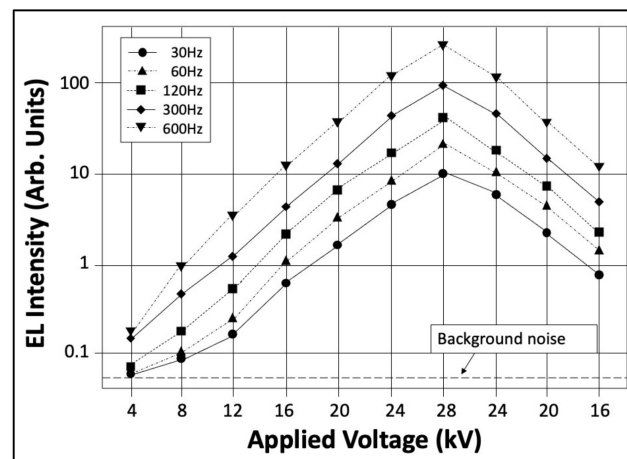


Figure 1. The electroluminescence intensity behaviour as a function of applied voltage. Reprinted with permission from ref. [26]. Copyright 2002 IEEE.

4.2. EL vs. Type of Materials

Different polymeric materials, such as LDPE, PET, XLPE, PP, and PEN, exhibit a similar behaviour of EL emission when subjected to AC voltage stresses. It is found that EL intensity in different materials rises with increasing voltage applied where light emission obtained is found to be homogenous. However, each material showed a different inception voltage for EL emission where the threshold voltage is 2.07 kV for PEN, 2.34 kV for PET and 2.76 kV for LDPE [8]. The EL intensity in the LDPE is substantially lower than in the PET and PEN samples. This is because both PET and PEN have an aromatic backbone in their polymer chains [8]. The aromatic backbone works as extra chromophores in the repeat units of the macromolecule that are coupled with the emitting luminescence centres, allowing for the production of more excited states [2].

A study by Kang Yang et al. [21] found that each material has a different surface trap density and LDPE has the largest surface trap density followed by PTFE and PP. When electrons are injected into the polymer through high voltage stress, the charges will have to occupy the deep traps of the material before occupying its shallow traps. Many charge carriers recombined at shallow traps. Some will be captured by trap centres. Therefore, for material with a large surface trap density, this will decrease the recombination of opposite charges in polymer, thus emitting less light as shown in Figure 2. The trap density of the material has a reverse relationship with its EL intensity.

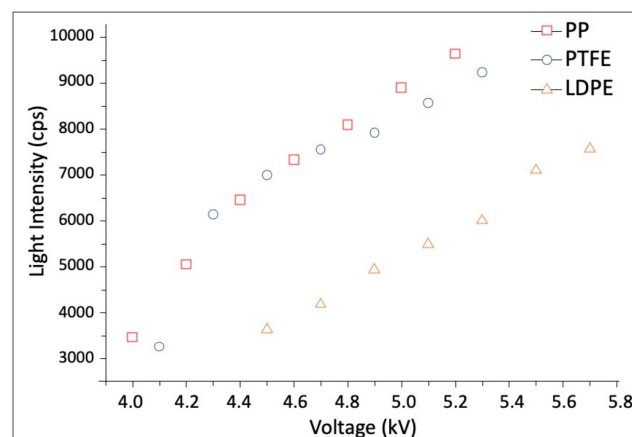


Figure 2. The EL scatter curves polymers vs. voltage. Reprinted with permission from ref. [21]. Copyright 2006 IEEE.

Another possible argument is that the EL intensity is influenced by the structure and the characteristics of the polymer itself. According to Shimizu et al., luminescence centres are connected with material crystallinity, hence crystalline polymers have greater brightness [27]. Light emission from polyethylene and PP are greater than that of polyvinyl chloride (PVC) and poly(methyl methacrylate) (PMMA). With its aromatic structure giving it strong luminescence properties and high field-withstanding capabilities, PEN is currently the most efficient insulating polymer in terms of electroluminescence yield [28]. When examining polyolefin polymer ageing mechanisms, a significant hurdle is the weak signal of EL intensity, making the understanding of recorded spectra a little challenging. The presence of a silver nanoparticles (AgNPs) layer on the side of the bi-oriented polypropylene (BOPP) film results in a significant increase in the EL signal, with the plasmon-induced electroluminescence exceeding three orders of magnitude [31].

4.3. EL vs. Presence of Water in Sample

Exposure of a polymer to strong electric stresses in the presence of water can lead to the formation of a water tree, which in turn can trigger the growth and initiation of electrical trees. Muramoto et al. [32] found that the electroluminescence characteristics can be influenced by the dried water-treed XLPE. The EL onset of dried water-treed XLPE is lower than that of virgin XLPE, whereas its EL intensity is higher, which enhances the quantity and size of micro voids and eventually boosts electron acceleration. This phenomenon is also related to the molecular structure of dried water-treed samples where free radicals' produced from electrons' collisions upon voltage application modified the polymer molecular structure and increased the presence of a luminescence centre within the polymer. Hence, it increases the chances of collision between electrons and luminescence centres [33].

4.4. EL vs. Mechanical and Electrical Ageing

In any high voltage cable that has operated for years, the ageing of material is bound to happen. There are two factors that can cause ageing of a polymeric material; the mechanical and the electrical factors [3]. In terms of mechanical factors, the effect of high external stresses causes mechanical stress which may induce sub-microcavities mechanically or weaken the bond strength of the polymeric material. It is generally accepted that at the microscopic scale, the formation of micro-voids will develop to a size large enough to sustain micro-discharges thus leading to the propagation of electrical treeing [30]. Meanwhile, the electrical factor is based on the influence of space charge [3]. During ageing, injected charges are trapped in the material and developed space charges. These space charges diminish the local field in the polymer while increasing the electric field at the semiconductor tip. This process significantly affects the number of injected charges and EL pulses produced in the aged polymer.

Ageing has a direct relationship with the excitation rate of luminescence centres because the space charge density and average mean free path carriers increase with the ageing duration [3]. The thermal and ultraviolet (UV) ageing of polypropylene films causes a reduction in EL emissions relative to that of unaged samples excited under similar experimental conditions [34]. Correspondingly, the EL intensity in XLPE cut from 77 kV class power cables and removed after 20 years of service gradually decreases with voltage application time and exhibits slightly different behaviour as compared to unaged XLPE. It is observed that the normalized EL intensity of the removed cable exhibits slow decreases in comparison to the unaged sample.

Additionally, the amount of luminescence centres and their species in XLPE cut from the removed cable are different from that of unaged XLPE [2,11]. According to Dissado et al., there is a proportionate relationship between the growth rate of trap density with age and the EL intensity [12]. The recombination of a mobile electron with a deep-trapped hole leads to the release of energy, which is then transmitted to the thermal motions

of the polymer molecules. This phenomenon has the potential to break down chemical bonds and consequently, increase the shallow and deep traps' density.

In [20], LDPE samples are subjected to thermal ageing in an oven for varying time intervals ranging from 120 h to 600 h. The EL intensity of all samples exhibits a lower EL inception voltage than the unaged sample. It is also found that the EL intensity of aged LDPE from 0 h to 360 h decreased with increasing ageing periods, but for 480 and 600 h, the EL intensity is larger than 360 h but still lower than 0 h as portrayed in Figure 3.

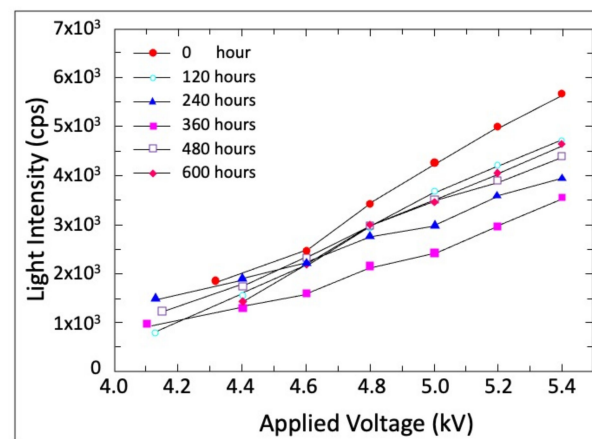


Figure 3. Relation between EL intensity of various aged LDPE samples and applied voltage. Reprinted with permission from ref. [20]. Copyright 2008 Elsevier.

The EL onset voltage of unaged samples are found to be higher than the aged sample and this is attributed to two primary reasons. Firstly, the oxidation process leads to an increase in surface traps, resulting in a lowering of the surface barrier of the LDPE sample, which impacts charge injection into the polymer. Secondly, the oxidation process increases the bulk traps in the material, which leads to a change in the space charge distribution, distortion of the electric field, and ultimately influences the charge injection [20]. The drop in EL is attributed to a decreased contact potential barrier following age, which facilitates charge injection near electrodes. Increased charge trapping and space charge buildup are also caused by higher bulk traps after ageing. This effect will reduce the recombination rate of trapped electrons and injected holes, hence lowering the EL intensity [20].

4.5. EL vs. Applied Frequency

The frequencies of applied voltage can have a very strong effect on the insulation of a polymeric cable. The appropriate frequency range for online diagnostic checks on underground power lines may be determined using the EL method [26]. A study by Bamji et al. [35] observed EL phenomena in unaged and aged XLPE insulation cables over a range of frequencies spanning from 5 Hz to 600 Hz. Their findings, as depicted in Figure 4, indicate that the amount of EL pulses emitted during positive and negative half-cycles in a given duration increases with rising frequency and voltage.

The injection of charges into the polymer is observed to increase with an increase in the frequency and voltage of the applied voltage. This increase in the mobility of injected charges causes them to spread out over a larger area from the semiconductor tip, leading to a larger EL zone and increased chances of EL emission. With an increase in frequency, the voltage remains above the EL emission threshold level for a shorter duration during each half-cycle. A similar result is obtained for studies conducted on LDPE [36] and commercial low molecular weight polybutadiene [37]. Therefore, the frequency range of 5 Hz to 600 Hz is recommended for on-site testing of underground power cables [35]. Two separate electrodes, i.e., Indium Tin Oxide (ITO) and gold (Au), were used to compare the efficiency of EL intensity on PEN. The sandwiched structures of ITO-PEN-ITO and Au-PEN-Au were subjected to up to 300 kV/mm at different frequencies under DC and

AC stresses. There was no notable distinction observed between the two types of electrodes under DC stress. Nonetheless, when subjected to AC stress, a visible embossment emerged in the low field region for the Au-PEN-Au sample, which grew more pronounced with increasing frequency, in contrast to the ITO-PEN-ITO sample [38,39]. The EL intensity increased with increasing stress frequency at the constant field. The same phenomenon was observed in LDPE when subjected to an applied voltage of 6 kV_{peak} at 10 Hz, 50 Hz and 100 Hz [40].

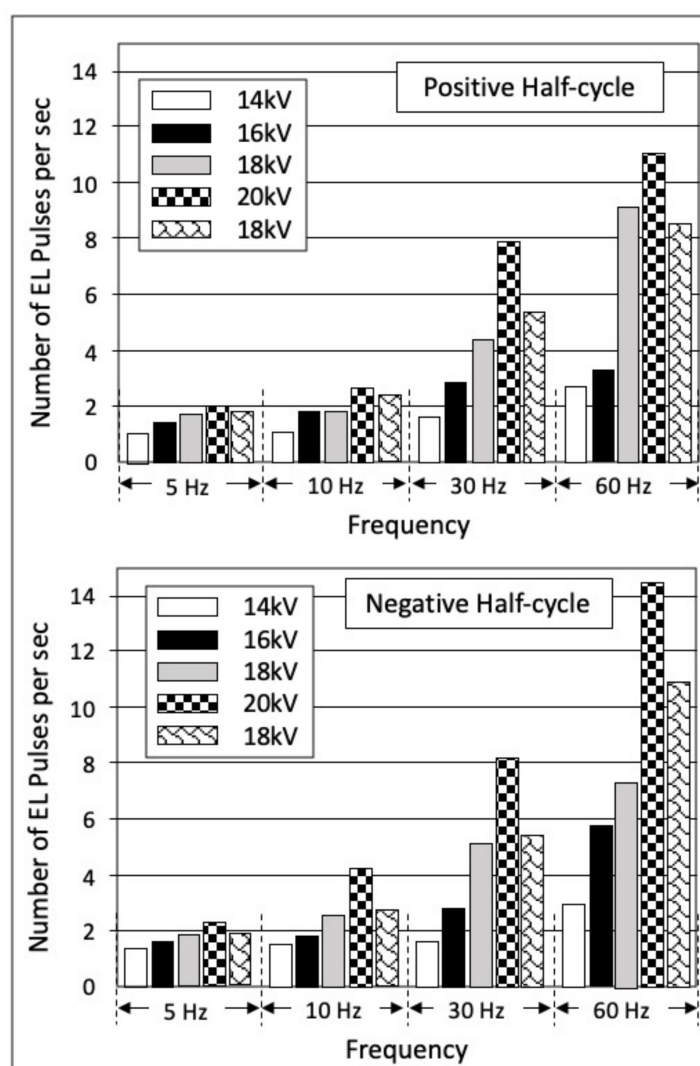


Figure 4. Relationship between the number of EL pulses per second and frequency across different voltages. Reprinted with permission from ref. [35]. Copyright 2001 IEEE.

Nevertheless, the number of pulses per cycle was found to be declining with increasing frequency [2,15,27,35,41]. This can be attributed to the fact that at each voltage cycle, the surface states will be filled by the injected charges before filling the bulk traps in the polymer. The electric field remains higher than injection threshold for an extended duration at low frequencies, allowing more time for the charges to migrate and get trapped in the polymer bulk. This results in enhanced recombination of opposite charges during the following half-cycle, leading to an increase in the intensity of EL emission [15].

4.6. EL vs. Time of Voltage Application

Shimizu et al. [10] found that the EL emission in LDPE increased initially with increasing time but subsequently decreased after reaching a peak value which varies across

different samples. The time of voltage application was between 0 min to 300 min. The application of voltage for an extended period of time can lead to changes in the molecular structure of the material, where the number of polar groups is increased. The same result was obtained in XLPE with removed cross-linking by products and oxygen [42]. It was found that the EL intensity increases gradually until 75 min before decreasing steadily with the time of voltage application up to 300 min.

The same phenomena are observed in dried water-treed XLPE at 5 kVrms [33]. However, at higher voltage applications, namely 7.5 kVrms or 10 kVrms, the light intensity will suddenly increase which shows the growth of tree channels as observed in Figure 5.

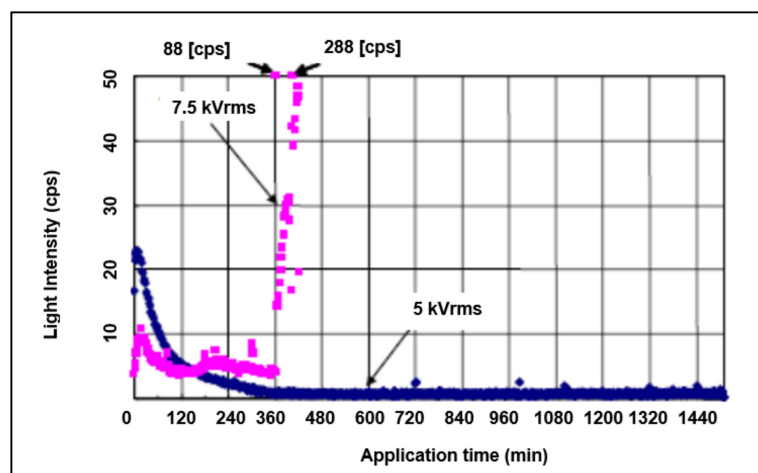


Figure 5. Light intensity (photon counts per second) vs. voltage application time at 5 kVrms and 7.5 kVrms [33].

The recombination of opposite polarity charge carriers in the bulk of the polymer is a critical factor affecting the electroluminescence emission. It is suggested that it takes a considerable amount of time for charge carriers to migrate from the electrodes into the bulk of the polymer [8]. During this process, the impacts between charges and molecules of the polymer can lead to molecular chain scission, resulting in changes in the energy level and concentration of the deep trap [27]. This causes the light intensity to decrease with increasing time application.

A study in [35] investigated the EL emission of XLPE when exposed to 15 kV (60 Hz) for 40 h. The EL intensity was observed to increase initially for half an hour before steadily decreasing with time [35]. According to Bamji et al., this scenario is attributed to the polynuclear aromatics of XLPE that introduce impurity molecules in polymer where they recombine with injected charges and produce EL emission during the initial application of voltage. However, less EL emission is detected as the molecules age because they are not re-ionised during impact collisions nor brought into an excitation state by the injected charge [35].

4.7. EL vs. Types of Voltage Waveform Applied

The types of waveform applied on polymer such as DC, sinusoidal AC, the trapezoidal and square waveform can have a great influence on the amount of EL intensity produced by the material. This effect is linked to the degree of charge carriers introduced into the polymer and the recombination of opposing charges. A great example is observed from the work of Bamji et al. [9]. The onset voltage of light emission of half-rectified AC (HRAC) is approximately twice the onset voltage for AC. EL intensity of both types of applied voltage increased with increasing voltage. Nonetheless, no emission of light was observed under DC stress up to the DC power supply's maximum of 46 kV. This is because the DC voltage application injected a unipolar charge into the polymer, which decreases the field at the injecting electrode, thereby preventing the occurrence of light emission or treeing.

As attempted by many researchers [5,9,41,43,44] despite the different experimental conditions and configurations, the EL intensity of polymeric material under AC stress has drawn the same conclusion; The applied sinusoidal voltage's EL is greatest in the first and third quadrants.

When an AC cycle is subjected to a high electric field, holes are injected during the positive half cycle and electrons are injected during the negative half cycle. These carriers get trapped in shallow and deep traps. The electrons will recombine with trapped holes in deep traps due to their inability to exit when the polarity is reversed. This recombination results in light emission. After recombination, the deep trap returns to its original condition [9]. Similar activity was observed for the subsequent positive half-cycle. Every AC voltage cycle will undergo the same process, resulting in electroluminescence emission at the first and third quadrant of the applied AC cycle. It is well-accepted that the intensity of electroluminescence emission during the positive half-cycle is lower than that of the negative half-cycle because electrons are much easier to inject than holes [43]. With more electron injection, more recombination with trapped holes can occur, therefore more emissions of light. Another study by Bamji et al. [43] on XLPE, reported the effect of electroluminescence properties when subjected to impulses superimposed on AC voltage. Impulses of ± 10 kV at 60 Hz AC, with a rise and fall time of 60 μ s and 750 μ s respectively, are superimposed at different phase angles [43]. The study revealed that the magnitude of EL intensity under the influence of impulse voltage was 50 times larger than normal AC voltage (without impulses) when the impulse voltage has the same polarity as the AC half-cycle. This implies that superimposing impulses with the same polarity as the half-cycle of the AC voltage increases the voltage above its inception level, resulting in charge injection into the polymer. Bamji et al. reported that the highest EL emissions were obtained at the peak of the AC cycle as the impulses raised the applied voltage to 30 kV [43]. Conversely, impulses with the opposite polarity of a half cycle of AC voltage, did not exhibit any effects, since they reduced the applied voltage to a level lower than the charge injection threshold.

Hinata et al. [13] reported the application of trapezoidal waveforms on magnesium oxide MgO/LDPE nanocomposite material. The EL intensity of this study, similar to other material exposed to AC voltage, increased shortly following the change in dE/dt to non-zero value and reached its maximum well before dE/dt reduced to its original value. Additionally, there were more EL occurrences in the $dE/dt = 0$ region than in the $dE/dt > 0$ region.

Another electroluminescence study was conducted on a thin LDPE sheet subjected to a uniform field configuration under AC stresses at room temperature [14]. Three types of voltage waveforms were applied to this material; sinusoidal AC, periodic square and triangular waveforms. The ΔEL in [14] represented the difference in intensity levels between the maximum and lowest for each waveform as shown in Figure 6. The square applied voltage has a greater EL intensity than the sinusoidal and triangular waveforms. This effect is due to the rapid changes of the square waveform from a positive to a negative applied voltage that leads to a higher amount of chargers injected into the insulation.

Distinct characteristics of the electroluminescence phenomenon were observed in sinusoidal AC, periodic square, and triangular excitation waveforms. Regardless of the excitation waveform used, the electroluminescence intensity during the positive voltage half cycle was observed to be 30% greater than that during the negative half cycle, even though the field strength at the injecting electrode was below the applied field. The difference was in the phase of the electroluminescence peak intensity concerning the applied field whereby the electroluminescence peak was leading in the sinusoidal waveform and was in phase in the triangular and square waveform. The EL intensity for square waveforms was much higher, about threefold in magnitude, than for sinusoidal and triangular waveforms [45].

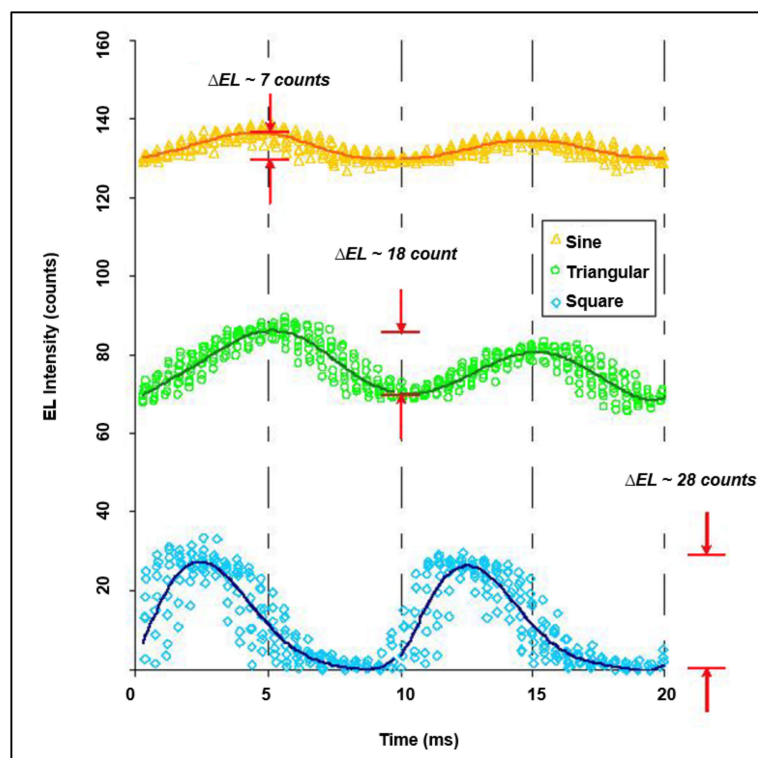


Figure 6. Comparing the EL intensity level for one cycle of sinusoidal, triangular and square ac voltages. Reprinted with permission from ref. [14]. Copyright 2008 IEEE.

4.8. EL vs. Influence of Various Gases

The amplitude intensity and threshold of light emission can be influenced by the presence of gases, depending on their electron affinity and chemical reactivity [17]. The initiation of electrical treeing for the degassed polymer takes longer than for normal polymer because a reduction in oxygen quantity leads to a decrease in UV radiation emission-induced photodegradation [9–11,46].

The EL inception voltage of polymer (needle-plane sample) increases for higher oxygen concentration because the electronegativity of oxygen causes electrons to attach with each other, thus reducing the recombination with opposite charges leading to an increase in inception voltage [44].

SF₆ and oxygen gas have a high electron affinity and can interfere with the light emission process by capturing the injected electrons and this will lead to lower light intensity counts [17]. The highly reactive nature of the gases indicates that the radical sites of the polymer chains are expected to interact with oxygen (O₂) and sulphur hexafluoride (SF₆) molecules, leading to the formation of more bond breaks [17,47,48]. The presence of oxygen in SF₆ gas can diffuse into the material and potentially lead to early tree initiation [10]. As reported by Bamji et al. [47], the EL inception voltage in normal samples with air in their free volume and in SF₆-impregnated samples is higher than in degassed polymer because of the electronegative nature of oxygen (in the air).

The EL intensity of nitrogen, N₂-impregnated polymer, does not exhibit a significant difference from degassed polymer [47,48]. N₂ is a chemically inert gas and has zero electron affinity [48]. Therefore, it will not react with free radicals. The EL onset voltage for N₂-impregnated polymer was found to be a little higher but with less EL intensity level. This is due to the fact that, under hot electron excitation, there is limited space for the injected electrons to gain enough kinetic energy from the electric field, resulting in fewer ionizations [49]. The photochemical reaction of the normal and degassed samples can be observed in [50]. The explanation of the photochemical reaction can be obtained elsewhere [50]. Generally, free radicals in the degassed polymer will produce inactive

products by recombining with each other. Conversely, free radicals in a normal sample will interact with oxygen or other polymer molecules, leading to further chemical reactions that accelerate the polymer degradation process.

The influence of surrounding conditions can have a substantial consequence on the EL emission characteristics. Polymer tested in pressurized gas was found to generate light emission from inside of the polymer (for needle-plane configuration) and those tested in vacuum were observed to emit light from the polymer surface [5]. In the pressurized gas, the mean free pathways of released electrons are relatively shorter, which prevents them from accumulating enough energy to induce photoluminescence. The amplitude and the amount of EL emission for the evacuated chamber are larger than observed in the polymer surface. This is because electron emission happened concurrently in both the sample and the vacuum, leading to a rise in the amplitude and number of EL pulses [51].

4.9. EL vs. Temperature

It is imperative to explore the properties of EL emission at high temperatures since most electrical engineering systems work at temperatures as high as 300 K. However, there are not many works of literature that report on the influence of temperature of working conditions on the intensity of EL emission. In contrast, several literatures are investigating the impact of temperature on the space charge distribution [52–54]. As described previously, space charges have a huge impact on the local field of polymer that consequently affect the intensity of EL emission. Furthermore, EL emission also depends on the rate of injection and migration of charges in polymer, the recombination rate of opposite charges and also the energy levels of the polymer where all abovementioned can be greatly affected by the working condition temperature.

Mary et al. [55] investigate the behaviour of EL features when subjected to varying temperature under AC and DC stress in a PEN sample. Both tests exhibit different EL features at below and above room temperature as can be seen in [55]. A similar result was obtained in [56] where the electroluminescence characteristic of PEN showed a clear dependency toward temperature, with discernible changes observed between low and room temperatures.

In AC stress, the EL emission during heating and cooling demonstrates a similar trend but with significant horizontal shift. At temperatures below room temperature, EL intensity decreases with increasing temperature, whereas it increases with temperature above room temperature. This phenomenon is presumed to be related to the growth in charge recombination rate at temperatures above ≈ 20 °C, which can be caused by the increase in the current injection at the electrodes or the number of effective charges trapped in the polymer [55]. Meanwhile, in DC stress, EL emission reaches its peak at -100 °C and $+40$ °C. This is due to charge traps' distribution in the polymer during heating and cooling. Charges experience a secondary relaxation process where the local motions of the chain segment influence the charge trapping and charge recombination rate [55]. More investigation on the effect of temperature on EL emission is required to explain the varying phenomenon scientifically.

5. EL Technique and Other Space Charge Measurement Methods

When exposed to high voltage, the measurement of electric stress in a dielectric material is critical in determining the material's breakdown behaviour. Polarisation happens when a uniform electric field is supplied across the dielectric material. Nevertheless, the existence of space charge produces non-linearity in the dielectric's local internal field, which affects the dielectric's behaviour [57]. It is well-accepted that electric stress is significantly affected by the space charge generation and accumulation in the dielectric, which could also lead to the breakdown of the material. Space charge can occur within a solid, liquid or gas dielectric.

Numerous researchers have examined the behaviour of space charges in great detail utilising nondestructive experimental methods, including the Laser Induced Pressure Pulse

Method (LIPP), the Pulsed Electro-Acoustic Method (PEA), the Thermal Step Method (TSM), and the Laser Intensity Modulation Method (LIMM). The primary goals of conducting space charge measurements are to measure the magnitude and orientation of charges under the influence of an electric field, calculating the depth and pattern of the charge layer, and comprehending the polarization mechanism and its impact on charge buildup [57]. However, each method has its own set of advantages and disadvantages.

This leads to a constant debate on which method is more reliable to investigate space charge distribution and detect the degradation of a material, especially when another potential technique, i.e., EL technique, attracts much attention. Several researchers [15,19,20,58] make some reliable comparisons between EL technique and other space charge measurement methods, and these converge to a conclusion; EL measurements, in addition to space charge approaches, may be used to evaluate the electrical deterioration and ageing of a polymer. Auge et al. [58] combined the EL technique and LIPP method to investigate PEN's internal space charge distribution under DC fields in the 25–225 kV/mm range. At low fields (50 kV/mm), EL is not seen, and the LIPP response indicates that the charge recorded at this field is a pure electrode charge. EL is detected at high electric fields (200 kV/mm) and increases substantially with field strength. The LIPP profiles provide clear evidence of charge injection into the bulk of the polymer, resulting in space charge generation [58].

To establish a strong relationship between EL and space charge, Le Roy et al. [19] employed a specially developed PEA device to simultaneously measure the space charge distribution and EL in LDPE under a uniform DC field. Although both methods yielded consistent results, it is essential to note that the detection of space charges only provided information on the net charge distribution, and that EL can be observed when opposing charges interact [19].

Bamji et al. [15] utilized a novel developed Phase-Resolved-Pulsed-Electro-Acoustic (PR-PEA) technique to examine the space charge distribution and electroluminescence (EL) in XLPE under an AC field at various frequencies from 1 Hz to 50 Hz. Again, the results obtained from both methods are consistent with each other. The study found that at lower frequencies, the charge migration and trapped space charges' accumulation in the polymer increased, which resulted in a higher rate of recombination of opposing charges and subsequent increase in EL intensity. This effect is closely connected to the behaviour of the space charge distribution, with the accumulation of more charges at 1 Hz than at 50 Hz as measured by the PR-PEA method [15].

Initially, Yang et al. [20] were unable to establish a definite relationship between space charge distribution and EL behaviours in aged LDPE under uniform AC stresses. However, through further investigation, they arrived at a similar conclusion as Bamji et al. [15] and Le Roy et al. [19]. It was found that during the ageing process, the bulk trap distribution within the material increased, causing more charges to be trapped and increase the accumulation of charges. This effect reduces the recombination rate of opposite charges, leading to lower EL intensity. Thus, a strong relationship between the distribution of space charge and EL phenomena was observed. Since light emission is highly dependent on radiative charge recombination, it also becomes one of the limitations in quantifying EL intensity. Therefore, a study in [56] suggested coupling thermally stimulated discharge current (TSDC) and thermally simulated luminescence (TL) as it can help to differentiate events related to charge detrapping.

6. Modelling of EL Using Dimensional Analysis

The injection and recombination of charge carriers of opposite polarities into the bulk of the polymeric substance are critical to the EL phenomena. It is also widely understood that EL can be indicative of electrical deterioration, and as a result, various alternative ageing and life models have been presented to forecast the onset of electrical failure. Various theories and explanations have been proposed to elucidate the mechanism of electrical degradation in insulating materials and the phenomenon of EL. One model simulates charge injection into a polymer from a metallic electrode under AC stress to

explain the emission phase. Another study also incorporated two bipolar charge models for simulating charge transport [30]. Serra et al. [59] developed a model to study the damage growth in insulation of an electrical cable under microscopic defects such as voids. The model simulated microcavities based upon the cavity size, electric field, and electron energy distribution. Meanwhile, Parpal et al. [60] developed a model that focused on the enlargement of nano- and mesocavities resulting from polymer rearrangements. Dissado and Wu [12], introduced a three-energy-level model to describe the sustained rise of the EL, portrayed by the growth of deep state concentrations, by considering the de-trapped rate of trapped charges. In contrast, Le Roy et al. [61] formulated a numerical model of charge transport to explain the mechanism of EL excitation by hot carriers and bipolar charge recombination.

An extensive electroluminescence model on LDPE has been proposed [40,45] that takes into account the charge carriers' recombination, injection, extraction and charge transport. The model produced a good correlation between measured and simulated phase-resolved electroluminescence intensity regarding its time dependency and types of applied voltage waveforms. For measurement of electroluminescence on DC bias voltage and AC voltage application after DC charging, the model showed the independence of electroluminescence intensity on DC bias voltage and the broken electroluminescence peaks during each half-cycle of the AC voltage after DC charging which supported the theory of dissymmetrical AC field applied after DC field.

The factors that influence the properties of EL in insulation have been discussed. By predicting the process of electrical ageing in insulation, any major catastrophe that may occur can be avoided. Therefore, it is important for an EL model to be developed in order to infer insulation ageing under AC stress with several other factors taken into consideration. Moving forward, from the authors' perspective, the Dimensional Analysis (DA) method can be opted to generate a mathematical relationship that can produce a reliable prediction for the electrical ageing process of polymeric insulation material.

DA is a powerful mathematical method for gathering knowledge on a phenomenon, such as EL, by utilizing dimensionally accurate equations [62]. One of the significant benefits of employing Dimensional Analysis is that it reduces the number of variables in a given set of equations. Numerous studies have employed DA to formulate plausible theories for complex physical scenarios. DA has been utilized in diverse applications such as determining the optimal design of an insulator in contaminated environments [63], establishing the correlation between flashover voltage and pollution resistance [64], predicting the life span of polymer tubes in solar hot water systems [65], scaling and measuring instrumented indentation [66], among others. In this work, some critical factors that impact the EL emission will be examined. The results will be modelled by applying DA mathematical outcomes and comparing them with experimental data from previous work.

The current paper investigates the influence of applied voltage, applied frequency, time of voltage application, material types and oxygen concentration on EL. Therefore, the relationship between these parameters can be expressed as follows;

$$ELI = (V, f, M, O_c, T) \quad (1)$$

where

ELI—electroluminescence intensity;

V—applied voltage;

f—applied frequency;

M—types of material;

O_c—oxygen concentration;

T—time of voltage application.

To present these parameters on a dimensional matrix, it is necessary to establish the appropriate physical units for each of them. For *ELI*, the charge recombination unit is used to represent EL intensity since they have a strong correlation between each other,

as explained before. If charge recombination increases, EL intensity also increases. For types of material used, the material depth trapped unit is employed since each material has different depth trapped and this influences the EL intensity. Material with larger depth trapped emits less EL. The units for these parameters are in length, mass, time and ampere (LMTA) system as shown in Table 1.

Table 1. Physical units for corresponding parameters used.

Parameters	Physical Units
Electroluminescence Intensity, <i>ELI</i>	$L^3T^{-2}A^{-1}$
Voltage, <i>V</i>	$L^2M^1T^{-3}A^{-1}$
Frequency, <i>f</i>	T^{-1}
Material trap depth, <i>M</i>	$L^2M^1T^{-2}$
Oxygen concentration, <i>O_c</i>	$L^{-3}M^1$
Time, <i>T</i>	T

The dimensional matrix of the above units can be represented as;

	<i>k</i> ₁ <i>ELI</i>	<i>k</i> ₂ <i>V</i>	<i>k</i> ₃ <i>f</i>	<i>k</i> ₄ <i>M</i>	<i>k</i> ₅ <i>O_c</i>	<i>k</i> ₆ <i>T</i>
L	3	2	0	2	-3	0
M	0	1	0	1	1	0
T	-2	-3	-1	-2	0	1
A	-1	-1	0	0	0	0

where *k*₁, *k*₂, *k*₃, *k*₄, *k*₅ and *k*₆ are the exponents of the parameters *ELI*, *V*, *f*, *M*, *O_c*, and *T*, respectively. The dimensional matrix rank is 4 and the dimensionless product of the whole set is 2 by deducting the number of variables with rank. The homogeneous linear equations with respect to the exponents of the parameters can be expressed as:

$$3k_1 + 2k_2 + 2k_4 - 3k_5 = 0 \tag{2}$$

$$k_2 + k_4 + k_5 = 0 \tag{3}$$

$$-2k_1 - 3k_2 - k_3 - 2k_4 = 0 \tag{4}$$

$$-k_1 - k_2 = 0 \tag{5}$$

The exponents *k*₃, *k*₄, *k*₅ and *k*₆ can be expressed in terms of *k*₁ and *k*₂ as follows:

$$k_4 = -\frac{3}{5} k_1 - k_2 \tag{6}$$

$$k_5 = \frac{3}{5} k_1 \tag{7}$$

$$k_6 = \frac{4}{5} k_1 + k_2 + k_3 \tag{8}$$

From Equation (8) we can see that the expression for *k*₆ is in terms of *k*₁, *k*₂ and *k*₃. Therefore *k*₃ is given the value of *n*. As there are two sets of dimensionless products; π_1 and π_2 , the solutions can be displayed in the matrix form as below;

	<i>k</i> ₁ <i>ELI</i>	<i>k</i> ₂ <i>V</i>	<i>k</i> ₃ <i>f</i>	<i>k</i> ₄ <i>M</i>	<i>k</i> ₅ <i>O_c</i>	<i>k</i> ₆ <i>T</i>
π_1	1	0	<i>n</i>	$-\frac{3}{5}$	$\frac{3}{5}$	$\frac{4}{5} + n$
π_2	0	1	<i>n</i>	-1	0	1 + <i>n</i>

Hence,

$$\pi_1 = \frac{ELI f^n O_c^{\frac{3}{5}} T^{\frac{4}{5}+n}}{d^{\frac{3}{5}}} \tag{9}$$

$$\pi_2 = \frac{V f^n T^{1+n}}{d} \tag{10}$$

By dimensional analysis,

$$\pi_1 = \phi(\pi_2) \tag{11}$$

Therefore,

$$\frac{ELI f^n O_c^{\frac{3}{5}} T^{\frac{4}{5}+n}}{d^{\frac{3}{5}}} = \phi\left(\frac{V f^n T^{1+n}}{d}\right) \tag{12}$$

By applying Buckingham’s π -theorem, the ELI in Equation (12) can be expressed as;

$$ELI = \frac{d^{\frac{3}{5}}}{f^n O_c^{\frac{3}{5}} T^{\frac{4}{5}+n}} \phi\left(\frac{V f^n T^{1+n}}{d}\right) \tag{13}$$

In this paper, only voltage and frequency parameters are considered. From previous discussions, the frequency f is approximately proportional to EL intensity. Therefore, from Equation (13), the ordinate is ELI , the abscissa is V and the parameter of the curve is $V \bullet f^n$ with T and d as constant to highlight the relationship between ELI and f . This resulted in,

$$ELI = k \phi(V f^n) \tag{14}$$

with k as a dimensional constant. Equation (14) will need to be constructed such that it will fit the graph from Figure 1 up to 28 kV. Since ELI vs. f exhibits an exponential curve, hence Equation (14) will have to take the form of,

$$ELI = k \cdot \exp(V f^n) \tag{15}$$

where k is between 0.01 and 0.03 by experiment. Meanwhile, the value of n is not fixed but changes with the amount of applied frequency. To fit the graph, n is denoted between -0.15 and -0.4 .

7. Modelling Results

From Equation (15), we can calculate the value of ELI and produce a graph to make a comparison with other researchers’ work. For $f = 60$ Hz, the value of k and n used for this graph are denoted as 0.01 and -0.31 respectively. The graph produced is shown in Figure 7. This graph is then compared with the results of Bamji et al., as shown in Figure 8. It can be observed that both experimental and model results exhibit a close relationship with each other.

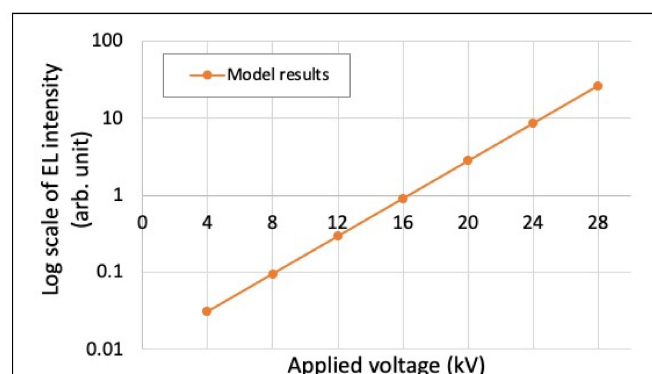


Figure 7. Log scale of EL intensity vs. applied voltage using model equation.

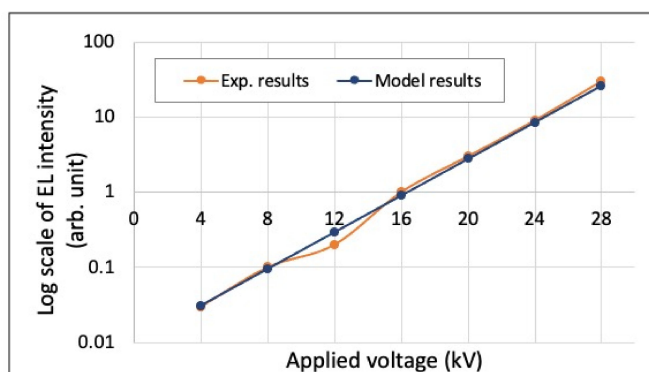


Figure 8. Log scale of EL intensity vs. applied voltage of experimental and model results.

However, for other frequency values, the value of k and n have to be changed in order to fit the graph as denoted in Table 2 below. It can be observed that the value of n increases as frequency increases and that the value of k is 0.01 for 30 Hz to 120 Hz but increases to 0.03 for larger frequency. The graph developed is as shown in Figure 9. Deeper investigation is needed to justify this condition.

Table 2. Parameters used to calculate EL.

f (Hz)	k	n
30	0.01	−0.4
60	0.01	−0.31
120	0.01	−0.24
300	0.03	−0.20
600	0.03	−0.15

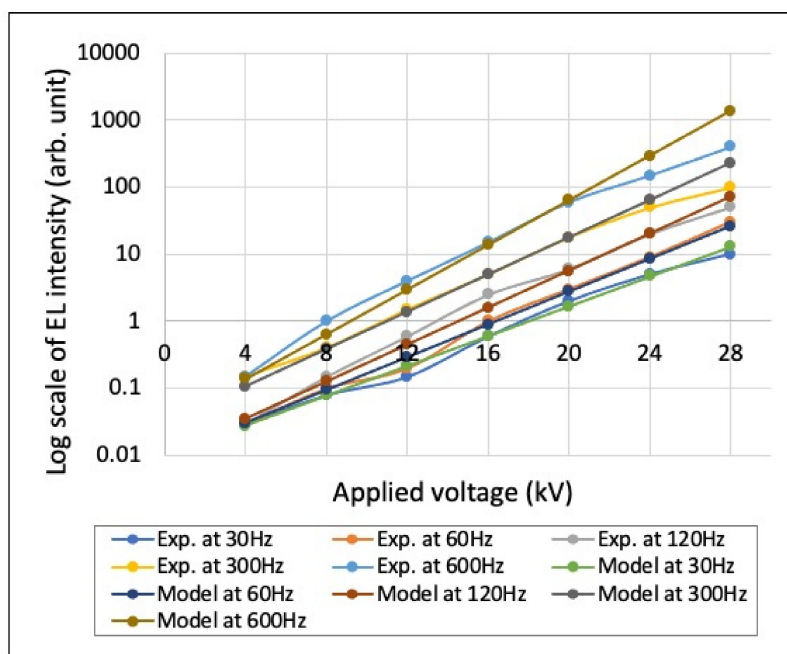


Figure 9. Log scale of EL intensity vs. applied voltage at various frequencies for experimental and model results.

8. Conclusions

This paper has outlined the main processes of the EL emission in polymeric material insulation. It is believed that the EL phenomenon arises from the interaction of injected and

trapped charges of opposite polarity within the polymer. The mechanism of EL emission necessitates excitation followed by de-excitation. Excitation states can be generated through impact ionization or bipolar recombination of opposite charges and the subsequent de-excitation will emit light upon returning to its ground state.

As discussed above, there are several advantages that can be obtained when employing EL techniques such as the possibility that it can be a good indication for the onset of electrical degradation even before the first PD occurs in the insulation; it has better performance than other commonly used partial discharge and space charge detection method by several orders of magnitude; it can observe the effect of space charge injection, decay and trapping in polymeric insulation when subjected to DC, AC, impulse voltages and other stress type; it is capable of detecting the transition from a water tree to an electrical tree which is impossible to obtain by other methods, and so forth.

Several strong factors have been discussed in order to understand the actual process that leads to the emission of light. From these factors, a mathematical relationship is crucial to be developed, and one of them is proposed using Dimensional Analysis in order to simulate the EL phenomena and predict the ageing of a polymer. Analysis of experimental and model data for *ELI* vs. *V* at various frequencies reveals a strong and consistent relationship. Additional research and experiments are required to verify the equation's validity for use with other frequency values.

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