

MEASUREMENT AND DIMENSIONAL ANALYSIS MODELLING OF
ELECTROLUMINESCENCE IN LOW DENSITY POLYETHYLENE MATERIAL

NURUL AINI BANI

UNIVERSITI TEKNOLOGI MALAYSIA

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NURUL AINI BINTI BANI

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ABSTRACT

Ageing process in power cables is inevitable especially cables under prolonged application of strong electric field for many years. Ageing is normally associated with space charge accumulation at the electrodes or in the bulk of the polymeric material. Many established non-destructive methods are available in detecting the space charge distribution in the material. However, most of these methods have limited spatial resolution thus producing large uncertainty in observing the space charge profiles near the electrodes. One method that has captured a lot of attention over the years for its reliable results and data thus enhancing the knowledge in the initiation of electrical ageing of polymeric insulation is known as electroluminescence (EL) method. EL is associated with the generation of charge carriers within the polymeric material and that these charges can be produced by injection, detrapping and field-dissociation. This thesis is based on the investigation of EL emission in virgin and aged low density polyethylene (LDPE) subjected to high alternating field. LDPE is chosen as the investigated material due to its vast usage in high voltage field. The behaviour of EL emission which can be affected by several factors such as, among others, applied voltage, applied frequency, ageing of material and types of materials and gases used. In all experiments, EL emission increases with increasing applied voltage. In aged LDPE, EL emission increases with increasing ageing temperature but decreases with increasing ageing duration. However, no specific pattern can be concluded for EL emission under applied frequency. A mathematical approach relating some of these factors and the intensity of EL is proposed and developed through the aid of Dimensional Analysis method. A close relationship between the model and experimental data obtained suggests that this mathematical approach can be utilized as a tool to predict electrical ageing of insulation material.

ABSTRAK

Proses penuaan dalam kabel kuasa tidak dapat dielakkan terutamanya kabel di bawah penggunaan medan elektrik yang kuat selama bertahun-tahun. Penuaan biasanya dikaitkan dengan pengumpulan caj ruang pada elektrod atau sebahagian besar daripada bahan polimer. Banyak kaedah bukan pemusnah boleh didapati bagi mengesan taburan caj ruang dalam bahan. Walau bagaimanapun, kebanyakan kaedah ini mempunyai resolusi ruang yang terhad, oleh itu menghasilkan ketidakpastian besar untuk pemerhatian caj ruang bagi profil berhampiran elektrod. Satu kaedah yang menarik banyak perhatian sejak beberapa tahun kerana keputusan yang boleh dipercayai dan data yang dapat meningkatkan pengetahuan kita dalam permulaan penuaan elektrik penebat polimer adalah dikenali sebagai kaedah elektropendarkilau (EL). EL dikaitkan dengan penjanaan pembawa caj dalam bahan polimer dan caj ini boleh dihasilkan melalui suntikan, penyingkiran perangkap dan penceraiian medan. Tesis ini adalah berdasarkan kepada kajian terhadap pelepasan EL dalam polietilena berketumpatan rendah (LDPE) yang dara dan tua yang dikenakan medan ulangalik tinggi. LDPE dipilih sebagai bahan disiasat disebabkan oleh penggunaan yang meluas dalam bidang voltan tinggi. Pelepasan EL boleh dipengaruhi oleh beberapa faktor seperti, antara lain, voltan yang diaplikasikan, frekuensi yang diaplikasikan, penuaan bahan, jenis bahan dan gas yang digunakan. Dalam semua ujikaji, pelepasan EL meningkat dengan peningkatan voltan yang diaplikasikan. Dalam LDPE yang tua, pelepasan EL meningkat dengan peningkatan suhu penuaan tetapi menurun dengan peningkatan masa penuaan. Walaubagaimanapun, tiada pola yang spesifik boleh disimpulkan untuk pelepasan EL di bawah frekuensi yang diaplikasikan. Pendekatan matematik berkaitan beberapa faktor ini dan keamatan EL telah dicadangkan dan dibangunkan melalui bantuan kaedah Analisis Dimensi. Hubungan yang rapat antara model dan data ujikaji yang diperolehi mencadangkan bahawa pendekatan matematik ini boleh digunakan sebagai alat untuk meramalkan penuaan elektrik bahan penebat.

TABLE OF CONTENTS

CHAPTER	TITLE	PAGE
	DECLARATION	ii
	ACKNOWLEDGEMENT	iii
	ABSTRACT	iv
	ABSTRAK	v
	TABLE OF CONTENTS	vi
	LIST OF TABLES	xi
	LIST OF FIGURES	xiv
	LIST OF ABBREVIATIONS	xxiii
	LIST OF SYMBOLS	xxv
	LIST OF APPENDICES	xxvii
1	INTRODUCTION	1
	1.1 Problem Statement	3
	1.2 Objectives	5
	1.3 Scope of Work	5
	1.4 Significant Contributions	7
	1.5 Outline of the Thesis	9
2	ELECTROLUMINESCENCE IN INSULATING POLYMER	11
	2.1 Polymers as Engineering Material	11
	2.2 Polyethylene	12
	2.2.1 Microstructure of Polyethylene	15
	2.3 Electrical Degradation of Cable Insulation	16
	2.3.1 Space Charge Distribution	18

2.3.2	Electroluminescence Phenomenon	20
2.3.3	The Energy Band Model	22
2.4	Space Charge Formation	25
2.4.1	Electrode-Dielectric Interface	26
2.4.2	Charge Transport and Injection Principles	27
2.5	Mechanism of Electroluminescence	30
2.5.1	Excitation Process	31
2.5.2	Relaxation Process	33
2.6	Measurement of Electroluminescence	34
2.6.1	Field Configuration	34
2.6.2	Sample Material	36
2.6.3	Detection System	37
2.7	Factors Affecting EL Intensity	39
2.7.1	Applied Electrical Field	39
2.7.2	Type of Materials	40
2.7.3	Presence of Water In Sample	42
2.7.4	Mechanical and Electrical Ageing	43
2.7.5	Applied Frequency	44
2.7.6	Time of Voltage Application	46
2.7.7	Types of Voltage Waveform Applied	47
2.7.8	Influence of Various Gases	52
2.7.9	Temperature of Surrounding	54
2.8	Electroluminescence Modelling	56
2.9	Principles of Dimensional Analysis	58
2.9.1	Physical Quantities	58
2.9.2	Dimensional and Dimensionless Quantity	59
2.9.3	Dimensional Homogeneity	60
2.9.4	Methods of Dimensional Analysis	62
2.10	Statistical Analysis in Insulating Material	63
2.10.1	Weibull Distribution	64
2.10.2	Generalized Extreme Value Distribution	65
2.10.3	Johnson S_B	65
2.10.4	Goodness-of-fit (GOF) Method	66

	2.10.4.1 Kolmogorov-Smirnov test	66
	2.10.4.2 Anderson-Darling test	67
	2.11 Summary	67
3	ELECTROLUMINESCENCE EXPERIMENTAL METHODOLOGY	69
	3.1 Sample Preparation	69
	3.2 Electroluminescence Experimental Setup	73
	3.2.1 Faraday Cage	75
	3.2.2 Vacuum Chamber	75
	3.2.3 Vacuum Pump System	77
	3.2.4 High Voltage Bushing	78
	3.2.5 Gaseous Valves and Tank	78
	3.2.6 Sample Holder	79
	3.2.7 Electron Multiplication Charge Coupled Device (EMCCD) Camera	80
	3.2.8 Trigger System	84
	3.2.9 WinSpec Spectroscopic Software	85
	3.3 Experimental Procedure	86
	3.3.1 Imaging of Light Emission	88
	3.3.2 Temporal Behaviour Measurement	88
	3.3.3 Breakdown Measurement	89
	3.3.4 Point On Wave Measurement	89
	3.4 Summary	92
4	EXPERIMENTAL RESULTS AND DISCUSSION	93
	4.1 Electroluminescence Imaging	93
	4.1.1 Spatial Distribution Analysis	99
	4.2 Temporal Behaviour Measurement	101
	4.3 Absorbance and Transmission	103
	4.3.1 Ultraviolet and Visible Spectrophotometer	104
	4.3.2 Fourier Transform Infrared Spectroscopy	109
	4.3.2.1 Finger print region	113

4.3.2.2	Carbonyl group	113
4.3.2.3	Hydroxyl group	114
4.4	Statistical Analysis on Sample Breakdown	114
4.4.1	Virgin LDPE	116
4.4.2	Aged LDPE at 310 K	118
4.4.3	Aged LDPE at 350 K	119
4.5	Electroluminescence Experimental Measurements	122
4.5.1	Voltage Dependency of EL Emission	122
4.5.1.1	Virgin LDPE	123
4.5.1.2	Temperature-aged LDPE	125
4.5.1.3	Time-aged LDPE	136
4.5.1.4	Discussion on the behaviour of EL emission in virgin and aged LDPE	145
4.5.2	Frequency Dependency of EL Measurement	146
4.5.2.1	Varying applied field at fixed frequency	146
4.5.2.2	Varying frequency at fixed applied field	156
4.6	Summary	169
5	ELECTROLUMINESCENCE MODEL	170
5.1	Development of EL Model	171
5.1.1	Charge Recombination and Charge Injection	171
5.1.2	Modelling of EL using Dimensional Analysis	173
5.2	Model Validation	178
5.2.1	Value of α_2 and α_3	178
5.2.2	Value of R_{eh} and n	181
5.2.3	Validation of Model through Regression	182
5.3	Comparison between Modelling and Experimental Results for Virgin LDPE	183
5.3.1	Voltage Dependency of EL Intensity	183
5.3.2	Frequency Dependency of EL Intensity	186
5.4	Comparison between Modelling and Experimental Results for Aged LDPE	191
5.4.1	Aging Temperature Dependency of EL Intensity	191

5.4.2	Aging Time Dependency of EL Intensity	197
5.5	Application of Model as a Diagnostic Tool	202
5.5.1	Comparison between Materials	202
5.5.2	Life-time Cable Prediction	204
5.5	Summary	206
6	CONCLUSIONS AND FUTURE WORK	208
6.1	Conclusions	208
6.2	Future Works	212
	REFERENCES	214
	Appendices A - E	230

LIST OF TABLES

TABLE NO.	TITLE	PAGE
2.1	Various polymers used for electroluminescence experiments	37
2.2	Base quantities using SI units	59
2.3	Derived quantities using SI units	59
2.4	Fundamental symbols for physical dimensions	60
3.1	Settings for cold sputtering method	72
4.1	Comparison of EL imaging for virgin and aged LDPE	98
4.2	The breakdown voltage value of virgin and aged LDPE samples	115
4.3	Statistical distribution ranking based on Anderson-Darling and Kolgomorov Smirnov for virgin LDPE	117
4.4	Statistical distribution ranking based on Anderson-Darling and Kolgomorov Smirnov for aged LDPE at 310 K	119
4.5	Statistical distribution ranking based on Anderson-Darling and Kolgomorov Smirnov for aged LDPE at 350 K	120
4.6	The range, majority and median of the breakdown voltage as measured in virgin and aged LDPE samples.	121
4.7	Phase angle difference of EL peak between applied field and virgin sample at first and third quadrant	125
4.8	Phase angle difference of EL peak between applied field and aged sample at first quadrant for different ageing temperature	132
4.9	Phase angle difference of EL peak between applied field and aged sample at third quadrant for different ageing temperature	132

4.10	Phase angle difference of EL peak between applied field and aged sample of different ageing temperature at first and third quadrant	135
4.11	Phase angle difference of EL peak between applied field and aged sample at first quadrant for different ageing time	141
4.12	Phase angle difference of EL peak between applied field and aged sample at third quadrant for different ageing time	141
4.13	Phase angle difference of EL peak between applied field and aged sample of different ageing time at first and third quadrant	144
4.14	Phase angle difference of EL peak between applied field and sample at first and third quadrant at 80 Hz for varying voltage	148
4.15	Phase angle difference of EL peak between applied field and sample at first and third quadrant at 50 Hz for varying voltage	150
4.16	Phase angle difference of EL peak between applied field and sample at first and third quadrant at 20 Hz for varying voltage	152
4.17	Phase angle difference of EL peak between applied field and sample at first and third quadrant at 5 kVp for varying frequency	159
4.18	Phase angle difference of EL peak between applied field and sample at first and third quadrant at 6 kVp for varying frequency	162
4.19	Phase angle difference of EL peak between applied field and sample at first and third quadrant at 7 kVp for varying frequency	165
5.1	Parameter and its physical units	174
5.2	Values for α_2 and α_3 and their respective final equation	179
5.3	Set of parameters and its value for voltage dependency model	184
5.4	Set of parameters and its value for frequency dependency model	187
5.5	Set of parameters and its value for ageing temperature dependency model	192

5.6	Variation of parameters used in ageing temperature dependency model	194
5.7	Set of parameters and its value for ageing time dependency model	197
5.8	Variation of parameters used in ageing time dependency model	199
5.9	Relative permittivity of insulating material at room temperature	202

LIST OF FIGURES

FIGURE NO.	TITLE	PAGE
2.1	The introduction of (a) crosslinking and (b) plastisizer agent can help increase the flexibility of the material	12
2.2	The formation of polyethylene from (a) methane gas which is converted to (b) ethylene before being heated and pressurized to become (c) polyethylene molecular chain	13
2.3	Structure of polyethylene	14
2.4	Formation of spherulite starts from (a) polymer crystals forming (b) crystal chains in a zig-zag conformation and these chains are folded upon each other to form (c) a stack of lamellae. The lamella consists of (d) an amorphous region (indicate by green curves) and a crystalline region (indicate by red curves). The lamellae grows to form (e) a spherulite	15
2.5	Possible defects within cable insulation	17
2.6	Failure in high voltage cable can caused by (a) large vented water tree growing from the insulation shield and (b) poor workmanship during installation process of cable insulation. Adopted from Buchholz (2004)	18
2.7	Energy band gaps for (a) insulator, (b) semiconductor and (c) conductor	22
2.8	Energy band diagram for a covalently-bonded crystal (Dissado and Fothergil, 1992)	24
2.9	Charge transport and injection mechanisms in polymer	28
2.10	The bipolar recombination model under the application of AC voltage as proposed by Alison et. al (1994). Figure is re-illustrated based on Ariffin et. al (2007a)	32
2.11	Electroluminescence configuration of (a) divergent and	

	(b) uniform field	35
2.12	Setup arrangement of (a) PMT system (Cao and Boggs, 2005), (b) CCD camera system (Ariffin, 2008) and (c) both PMT and CCD camera system (Bamji and Bulinski, 2002)	38
2.13	Intensity of EL emission during 100 s as a function of applied voltage at various fixed frequencies. Adopted from Cisse et. al (2002a)	40
2.14	The EL scatter curves polymers as a function of voltage. Adopted from Yang et. al (2006)	41
2.15	Light intensity of virgin and dried water-treed XLPE as a function of applied voltage. Adopted from Muramoto et. al (2008)	42
2.16	Number of EL pulses per second as a function of the frequency at various voltages. Adopted from Bamji et. al (2001b)	44
2.17	Number of EL pulses per cycle as a function of the frequency at various voltages. Adopted from Bamji et. al (2001b)	45
2.18	Light intensity (photon counts per second) vs. voltage application time at 5 kVrms and 7.5 kVrms. Adopted from Muramoto et. al (2008)	46
2.19	Temporal behaviour of EL emission from XLPE during aging at 15 kV (60 Hz). Adopted from Bamji et. al (2001b)	47
2.20	Light intensity from degassed LDPE vs. the amplitude of AC, HRAC and DC voltages. All voltages are peak values. Adopted from Bamji et. al (1989)	48
2.21	The light emitted during negative half-cycle has higher intensity than during positive half-cycle by (a) Krause et. al (1990), (b) Bamji et. al (2001a)	49
2.22	Applied electric field waveform $E(t)$ and time distributions of number of EL under trapezoidal waveform application for LDPE/MgO 70 μm film (1 phr, NP sample). Adopted from Hinata et. al (2007)	50
2.23	Comparing the EL intensity level for one cycle of sinusoidal, triangular and square ac voltages. Adopted from Ariffin et. al (2008)	51

2.24	Light intensity as a function of applied high voltage with different radius of needle tip and oxygen concentration. Adopted from Krause et. al (1990)	52
2.25	Photochemical reactions in XLPE. Adopted from Bamji and Bulinski (1997)	53
2.26	Temperature dependence of the EL under 106 kV/mm crest AC field for thick film PEN. a, c, b and d are consecutive temperature ramp up (a,b) and down (c,d) at 5 °C/min. Adopted from Mary et. al (2002)	55
2.27	Temperature dependence of the EL of a thick film PEN under 280 kVmm ⁻¹ DC field. Sample is heated to 100 °C (a) before cooled down to -150 °C (b) for first cycle and heat up again to 100 °C (c) before finally cool down to -150 °C (d). Adopted from Mary et. al (2002)	55
3.1	The gold sputter in used is Emitech K550X coater	70
3.2	Schematic diagram for gold coating using cold sputtering method	71
3.3	The finished LDPE sample ready to be tested	73
3.4	The overall setup for the EL experiments	74
3.5	Schematic diagram of EL experimental setup	74
3.6	The Faraday cage with locking system	75
3.7	Vacuum chamber located inside the Faraday cage	76
3.8	(a) Vacuum pump connected to valve and (b) vacuum pump machine	77
3.9	(a) Active gauge controller and (b) pressure detector	77
3.10	(a) HV bushing inside chamber and (b) HV bushing outside chamber	78
3.11	(a) Nitrogen gas pipeline entry and (b) nitrogen gas tank	79
3.12	(a) Sample holder consists of two electrodes and (b) paper cover with a square window	80
3.13	The EMCCD used in the EL experiments	81
3.14	CCD camera operates on the principle of charge coupling	82
3.15	EMCCD extended multiplication register configuration	83

3.16	Camera trigger	85
3.17	Winspec software interface	86
3.18	Sample holder	87
3.19	Principle of point-on-wave measurement. Adopted from Chong et. al (2006)	90
3.20	Flow chart of experimental methodology for EL measurements	91
4.1	Sample produces good EL image intensity (a) before measurement at 4 kVrms and (b) after measurement at 8 kVrms	94
4.2	Sample with low EL intensity at 4 kVrms	94
4.3	Presence of bright spots indicate existence of unwanted particles	95
4.4	Breakdown starts from edge of sample	96
4.5	Breakdown due to broken gold layer	96
4.6	Breakdown sample with enlarged image of breakdown area	97
4.7	EL image of (a) virgin LDPE sample, (b) aged LDPE sample at 310 K, (c) aged LDPE sample at 330 K and (d) aged LDPE sample at 350 K	99
4.8	The comparison of emitted light intensity in virgin and aged samples.	100
4.9	Temporal behaviour measurement of virgin LDPE at (a) 425 minutes before breakdown and (b) 64 hours of complete testing	102
4.10	Overall ultraviolet and visible spectroscopy results for one-side gold coated samples	104
4.11	Transmittance measurement of virgin and aged LDPE samples at different ageing temperature for (a) uncoated films and (b) coated films on one side with ~20 nm gold layer	106
4.12	Transmittance measurement of virgin and aged LDPE samples at different ageing time for (a) uncoated films and (b) coated films on one side with ~20 nm gold layer	107
4.13	Shift in the wavelength of the transmission maximum	

	for increasing ageing temperature and time	108
4.14	Transmission measurement of virgin and aged LDPE samples at different ageing temperature	110
4.15	Transmission measurement of virgin and aged LDPE samples at different ageing days	110
4.16	Oxidation reactions occurred in LDPE	111
4.17	Transmission measurement of virgin and aged LDPE samples at different ageing temperature for (a) hydroxyl and (b) carbonyl group	112
4.18	Transmission measurement of virgin and aged LDPE samples at different ageing days for (a) hydroxyl and (b) carbonyl group	112
4.19	Probability density function for virgin LDPE	116
4.20	Probability density function for aged LDPE at 310 K for 3 days.	118
4.21	Probability density function for aged LDPE at 350 K for 3 days	120
4.22	POW measurement for virgin sample subjected to applied field of 3 kVp to 7 kVp at a constant 50 Hz, AC sinewave	123
4.23	Phase angle position of EL peak between applied field and virgin sample at first and third quadrant	124
4.24	POW measurements for sample aged at (a) 310 K, (b) 330 K and (c) 350 K subjected to applied field of 3 kVp to 7 kVp at a constant 50 Hz, AC sinewave	126
4.25	Peak of EL intensity at each voltage level for first and third quadrant for sample aged at (a) 310 K, (b) 330 K and (c) 350 K	128
4.26	EL intensity peak at (a) first quadrant and (b) third quadrant for all voltage at all temperature	129
4.27	Phase angle position of EL peak between applied field and aged sample at (a) first quadrant and (b) third quadrant	130
4.28	Comparison of virgin and thermally aged LDPE at 7 kVp of applied field with constant 50 Hz, AC sinewave	133

4.29	Comparison of phase angle of virgin and aged LDPE at 7 kVp for (a) first quadrant and (b) third quadrant	134
4.30	Average value of EL intensity for virgin and aged sample subjected to applied field of 3 kVp to 7 kVp at a constant frequency of 50 Hz, AC sinewave	135
4.31	POW measurements for sample aged at (a) 6 days, (b) 12 days and (c) 21 days subjected to applied field of 3 kVp to 7 kVp at a constant 50 Hz, AC sinewave	136
4.32	Peak of EL intensity at each voltage level for first and third quadrant for sample aged at (a) 6 days, (b) 12 days and (c) 21 days	138
4.33	EL intensity peak at (a) first quadrant and (b) third quadrant for all voltage at all aging days	139
4.34	Phase angle position of EL peak between applied field and aged sample at (a) first and (b) third quadrant for different ageing time	140
4.35	Comparison of virgin and timely aged LDPE at 7 kVp of applied field with constant 50 Hz, AC sinewave	142
4.36	Comparison of phase angle of virgin and timely aged LDPE at 7 kVp for (a) first quadrant and (b) third quadrant	143
4.37	Average value of EL intensity for virgin and aged sample subjected to applied field of 3 kVp to 7 kVp at a constant frequency of 50 Hz, AC sinewave at different ageing days	144
4.38	POW measurement for virgin sample subjected to applied field of 3 kVp to 6 kVp at constant frequency of 80 Hz, AC sinewave	147
4.39	Peak of EL intensity at each voltage level for first and third quadrant at constant frequency of 80 Hz, AC sinewave	174
4.40	Phase angle position of EL peak between applied field and sample at first and third quadrant at 80 Hz	148
4.41	POW measurement for virgin sample subjected to applied field of 3 kVp to 6 kVp at constant frequency of 50 Hz, AC sinewave	149
4.42	Peak of EL intensity at each voltage level for first and third quadrant at constant frequency of 50 Hz, AC	

	sinewave	149
4.43	Phase angle position of EL peak between applied field and sample at first and third quadrant at 50 Hz	150
4.44	POW measurement for virgin sample subjected to applied field of 3 kVp to 6 kVp at constant frequency of 20 Hz, AC sinewave	151
4.45	Peak of EL intensity at each voltage level for first and third quadrant at constant frequency of 20 Hz, AC sinewave	151
4.46	Phase angle position of EL peak between applied field and sample at first and third quadrant at 20 Hz	152
4.47	Phase angle position of EL peak between applied field and all samples at first and third quadrant for all frequency	153
4.48	Peak of EL intensity at each voltage level for (a) first quadrant and (b) third quadrant for all samples	154
4.49	The average value of EL intensity for applied frequency of 20 Hz, 50 Hz and 80 Hz at all voltage steps	155
4.50	POW measurement for virgin sample subjected to varying frequency of 10 Hz to 100 Hz at constant field of 5 kVp	157
4.51	Peak of EL intensity at each frequency level for first and third quadrant at constant applied field of 5 kVp	157
4.52	Phase angle position of EL peak between applied field and sample at (a) first and (b) third quadrant at 5 kVp	158
4.53	POW measurement for virgin sample subjected to varying frequency of 10 Hz to 100 Hz at constant field of 6 kVp	160
4.54	Peak of EL intensity at each frequency level for first and third quadrant at constant applied field of 6 kVp	160
4.55	Phase angle position of EL peak between applied field and sample at (a) first and (b) third quadrant at 6 kVp	161
4.56	POW measurement for virgin sample subjected to varying frequency of 10 Hz to 100 Hz at constant field of 7 kVp	163
4.57	Peak of EL intensity at each frequency level for first and	

	third quadrant at constant applied field of 7 kVp	163
4.58	Phase angle position of EL peak between applied field and sample at (a) first and (b) third quadrant at 7 kVp	164
4.59	The average value of EL intensity for applied field of 5 kVp, 6 kVp and 7 kVp at all frequencies	166
4.60	Peak of EL intensity for applied field of 5 kVp, 6 kVp and 7 kVp at all frequencies for (a) first and (b) third quadrant	167
4.61	Phase angle position of EL peak between applied field and sample at all frequencies for (a) first and (b) third quadrant for all fixed applied voltage	168
5.1	Graph plots of EL modelling for (a) $\alpha_2 = 1$ and $\alpha_3 = 0$, (b) $\alpha_2 = 0$ and $\alpha_3 = 1$, (c) $\alpha_2 = 1$ and $\alpha_3 = 1$, (d) $\alpha_2 = 2$ and $\alpha_3 = 0$, (e) $\alpha_2 = 0$ and $\alpha_3 = 2$, (f) $\alpha_2 = 2$ and $\alpha_3 = 2$, (g) $\alpha_2 = 1$ and $\alpha_3 = 1$ with exponential function and (h) $\alpha_2 = 2$ and $\alpha_3 = 2$ with exponential function	180
5.2	The value of recombination density of mobile and trapped charges, R_{eh} with increasing value of n where $-10 > n > 10$ at applied field of 5 kVp	181
5.3	The value of recombination density of mobile and trapped charges, R_{eh} with increasing applied voltage from 0 kVp to 10 kVp at (a) $n = 2$ and (b) $n = -2$	182
5.4	Simulation of EL measurement with varying electrical field	184
5.5	Simulation of EL measurement (with 10% standard error) for virgin LDPE and compared with experimental data	185
5.6	(a) Best fitted line plot and (b) normal probability plot of virgin LDPE	186
5.7	Simulation of EL measurement with varying electrical field at applied frequency of 20 Hz, 50 Hz and 80 Hz	187
5.8	Simulation of EL measurement (with 10% standard error) for virgin LDPE and compared with experimental data at 20 Hz, 50 Hz and 80 Hz	188
5.9	Fitted line plot of simulated versus measured EL intensity at (a) 20 Hz, (b) 50 Hz and (c) 80 Hz	189
5.10	Normal probability plot of simulated versus measured EL	

	intensity at (a) 20 Hz, (b) 50 Hz and (c) 80 Hz	190
5.11	Simulation of EL measurement with varying electrical field at 310 K until 390 K	192
5.12	Simulation of EL measurement (with 10% standard error) for aged LDPE and compared with experimental data at 310 K, 330 K, and 350 K	193
5.13	Fitted line plot of simulated versus measured EL intensity at (a) 310 K, (b) 330 K and (c) 350 K	195
5.14	Normal probability plot of simulated versus measured EL intensity at (a) 310 K, (b) 330 K and (c) 350 K	196
5.15	Simulation of EL measurement with varying electrical field at 6 days until 24 days	198
5.16	Simulation of EL measurement with varying electrical field at 12 days until 24 days	198
5.17	Simulation of EL measurement (with 10% standard error) for aged LDPE and compared with experimental data at 6, 12 and 21 days	199
5.18	Fitted line plot of simulated versus measured EL intensity at (a) 6 days, (b) 12 days and (c) 21 days	200
5.19	Normal probability plot of simulated versus measured EL intensity at (a) 6 days, (b) 12 days and (c) 21 days	201
5.20	Simulated EL measurements for different ϵ_r of material with respect to increasing applied voltage	202
5.21	Simulated time to breakdown in days for material aged at 310 K, 330 K and 350 K with respect to varying applied voltage from 0 kVp to 10 kVp	204
5.22	Estimation of breakdown time for aged sample	205

LIST OF ABBREVIATIONS

A310, A330, A350	-	Aged LDPE at 310 K, 330 K and 350 K
A6, A12, A21	-	Aged LDPE at 6, 12 and 21 days
AC	-	Alternating current
AD	-	Anderson-Darling
CGS	-	System using centimetre, gram and second
DA	-	Dimensional analysis
EL	-	Electroluminescence
EMCCD	-	Electron Multiplying Charge-coupled device
FT-IR	-	Fourier transform infra-red
GEV	-	Generalized extreme value distribution
GOF	-	Goodness-of-fit
HDPE	-	High density polyethylene
HRAC	-	Half-rectified AC
KS	-	Kolgomorov Smirnov
LDPE	-	Low-density polyethylene
LIMM	-	Laser induced modulation method
LIPP	-	Laser induced pressure pulse
MKS	-	System using meter, kilogram and second
MLE	-	Maximum likelihood estimation
N ₂	-	Nitrogen
PE	-	Polyethylene
PEA	-	Pulsed electro acoustic
PEI	-	Polyetherimide
PEN	-	Polyethylene naphthalate
PES	-	Polyethersulfone

PET	-	Polyethylene Terephthalate
PI	-	Polyimide
PMMA	-	Polymethyl methacrylate
PMT	-	Photomultiplier tubes
POW	-	Point-on-wave measurement
PP	-	Polypropylene
PR-PEA	-	Phase-Resolved-Pulsed-Electro-Acoustic
PTFE	-	Polytetrafluoroethylene
PVC	-	Polyvinyl chloride
PWP	-	Pressure wave propagation
SF ₆	-	Sulfur hexafluoride
SI	-	System of Units
TPM	-	Thermal pulse method
TSM	-	Thermal step method
UV/Vis	-	Ultraviolet and visible spectrometer
XLPE	-	Crosslinked polyethylene

LIST OF SYMBOLS

A	-	Richardson-Dushman constant ($1.20 \times 10^6 \text{ Am}^{-2}\text{K}^{-2}$),
D_{c1}, D_{c2}, D_{c3}	-	Dimensionless constant
E	-	Electric field
E_g	-	Energy enforced on the valence electrons
E_f	-	Fermi energy
eV	-	Electron volts
h	-	Planck constant ($4.14 \times 10^{-15} \text{ eVs}$)
\hbar	-	$h/2\pi$
j	-	Current density
$k, \sigma, \mu, \lambda, \xi$	-	Continuous parameters
k_B	-	Boltzmann constant ($8.62 \times 10^{-5} \text{ eV K}^{-1}$)
m	-	Effective mass of electron ($9.11 \times 10^{-31} \text{ kg}$)
$M_{em,ht}$	-	Recombination coefficient for a mobile electron with a trapped hole
$M_{hm,et}$	-	Recombination coefficient for a mobile hole with a trapped electron
P	-	Probability
q_h, q_e	-	Charge of a hole and electron ($\pm 1.6 \times 10^{-19} \text{ C}$)
R^2	-	R-squared
R_{eh}	-	Recombination rate of charges
$R_{em,ht}$	-	Recombination of a mobile electron with a trapped hole
$R_{hm,et}$	-	Recombination of a mobile hole with a trapped electron
T	-	Temperature
T_g	-	Glass transition temperature
V	-	Applied voltage
X	-	Thickness of the sample

XY	-	Neutral molecule
$XY\cdot$	-	Excited molecule
XY_+	-	Ionized molecule
XY_0, VW_0	-	Molecular ground states
α, β, n	-	Constants parameters
α_2, α_3	-	Dimensionless group indices
ΔE	-	Difference between the maximum and minimum intensity levels for each waveform
\bar{e}_{hot}	-	Hot electron
\bar{e}_{th}	-	Thermalized electron
ϵ_0	-	Permittivity in a vacuum
π_1, π_2, π_3	-	Dimensionless group
ρ	-	Space charge density
ρ_{em}	-	Charge density of mobile electrons
ρ_{et}	-	Charge density of trapped electrons
ρ_{hm}	-	Charge density of mobile holes
ρ_{ht}	-	Charge density of trapped holes
φ	-	Work function
ϕ_{aged}	-	Phase angle difference of EL peak between aged sample and applied peak
ϕ_{diff}	-	Phase angle difference of EL peak between virgin and applied peak
$\phi_{diff-appliedfield}$	-	Phase difference of EL peak between virgin and aged sample
$\phi_{diff-virgin}$	-	Phase difference of EL peak between virgin and aged sample
ω	-	Applied angular frequency
$1,3XY\cdot$	-	First excited singlet or triplet state
$1,3h\nu$	-	Photon energy radiated by fluorescence or phosphorescence

LIST OF APPENDICES

APPENDIX	TITLE	PAGE
A	List of publications	230
B	Basic specifications of EMCCD camera	232
C	WinSpec software settings	235
D	List of statistical distribution ranking for all samples	239
E	Series of EL graph with different values of α_2 and α_3	243

CHAPTER 1

INTRODUCTION

Overhead lines and underground cables are two main forms of electrical energy transmission. Both types offer great advantages as well as disadvantages to the consumer and power distributor companies. The cost of manufacturing for underground cables is much higher than overhead lines. Underground cables are usually buried in the ground thus need to be insulated entirely whereas overhead lines take advantage of the convection of the ambient air to create some form of insulation. The system faults in underground cables are hard to detect and repair which increases the installation and maintenance cost.

Nevertheless, these do not stop the increase usage of underground cables. Several factors that influence the demand for underground cables are the reliability of service, environment conditions and impact, and restricted access among others. Overhead lines are predicted to be out of service much faster than underground cable because they produced outages more frequent than the underground cables. Underground cables are much less susceptible to environment conditions such as lightning strikes, air-borne pollutants as well as the potential of bird contacts. This eliminates the need for repeating maintenance operations as required by overhead lines that may increase the overall cost of operating overhead lines. For areas with restricted access for power transmission line such as airports and congested urban regions, it will be more convenient to use buried underground cables. Underground cables have a much lower series inductance than overhead lines because the high distance between conductor and earth. In addition to that, underground cables also

have a correspondingly higher capacitance, higher charging current and highly reduced resistance (Van Hertem *et al*, 2016).

Dielectric material acts as an insulation material for current-carrying electrical and electronic devices to protect them from high voltages. Dielectric materials are not only used in power apparatus but also in microcircuits. A good dielectric material required low permittivity, low dielectric loss, low conductivity and high dielectric breakdown strength. Dielectric material comes in different forms; solids such as ceramic, polyethylene, glass and silica, liquids such as silicone oil and transformer oil, gaseous such as air and SF₆ and also in vacuum form.

The vast majority of the current generation of high voltage cables uses insulation systems based upon polymeric material. There are several types of polymeric insulating material and the choice of this material is based upon their ability to meet certain key requirements for insulation systems. These requirements include high electric strength, low loss, reasonable flexibility, thermomechanically stable and economically attractive (Morgen E *et.al*, 2000). There are several other factors that need to be considered when choosing insulation material such as the nature of the polymer itself, the use of any additives and the manufacturing process (Ebewele, 2000). This is because different polymer exhibits different characteristics.

However, any polymers when subjected to prolong electric, mechanical, chemical, thermal and environmental stresses will sooner or later become deteriorate. In electrical degradation, discharges due to insulation voids, cavities, contaminants and protrusions act as a point where electric field is enhanced thus causing erosion to the material or forming conducting tracks in the system (Bamji *et. al*, 1989) It is crucial to predict the life of a cable in order to avoid catastrophic failure. Therefore, it is necessary to understand the optical, electrical and chemical properties of polymer as well as the nature of charge transport and migration in the polymer that contributes to the degradation of the material. One such method that can be used to detect early electrical ageing especially at interfacial region is known as electroluminescence (EL) method. Electroluminescence is the emission of light that

is related to successive excitation and de-excitation of valence electrons of polymer molecules upon application of high electric field.

1.1 Problem Statement

Under the prolong application of a strong electric field or after decades of operating life, the insulating material of a high voltage cable may experience degradation and ageing that can cause catastrophic breakdown. Ageing process comes in many forms such as thermal ageing due to temperature fluctuation, mechanical ageing due to vibration and bending, chemical ageing due to molecular changes in the bulk of polymer and many more. During aging, space charges are formed when injected charges are trapped in the trapping levels of polymer. The space charges enhanced the electric stress at the semicon tip and reduce the local field in polymer. Space charge is detectable through a range of non destructive methods such as the thermal pulse method (TPM), the thermal step method (TSM), the laser induced modulation method (LIMM), the laser induced pressure pulse (LIPP) and the pulsed electro acoustic (PEA) techniques. Each method differ in term of the way the internal space charge is perturbed which will produce a time dependent signal that allows the internal space charge distribution within the material to be detected by external circuit.

However, there are some limitations observed for most of these methods. In general, the limited spatial resolution of most space charge measurements (typically 10 μm) created uncertainty in the observing of the space charge profiles near the electrodes. In addition, most of these methods are based on the relative displacement of the space charge in respect to the electrodes thus modifying the influence of the charge on the electrodes. Moreover, the form and the evolution of the perturbation as a function of time are important elements that need to be identified during the measurements. PEA technique is the only technique that does not impose a displacement of charges in measuring the space charge distribution. However, PEA requires matching acoustic impedance for comprehensible signals to be detected. LIPP on the other hand provides poor data acquisition due to interference from

Pockels cell that it opted. LIPP method also requires appropriate pressure pulse to be targeted to the sample and the sample holder has to be strong enough to withhold several laser shots. The thermal methods which include TSM, TPM and L IMM are mostly suitable for thick samples of more than 2 mm thickness.

Another non-destructive method that is capable of investigating the interaction of charges within a polymer is the emission of light due to external electric field known as electroluminescence (EL). The light emission occurs in the visible spectrum before the onset of degradation mechanism such as electrical treeing and partial discharges. Under the application of AC electrical field, due to the nature of the positive and negative half cycle of the AC field, the injected charges are not able to migrate far into the bulk of the polymer but instead the charges will remain in close proximity to the injecting electrode during one half cycle before recombining with opposite polarity charge during the following half cycle. This enhances the robustness of EL method in investigating the interaction of charge within a very small region near the electrode-polymer interface. Moreover, EL method can be applied to thin film polymers ($< 100 \mu\text{m}$). The use of electron multiplying charged coupled device (EMCCD) cameras in the EL configuration system compensates the lack of spatial resolution in most space charge measurements thus offering uniformity in data acquisition. It is thought that the EL measurements may provide an alternative method to investigate the electrical ageing and degradation of polymer besides space charge probing. One of the main characteristics of EL is the variation of EL intensities with respect to insulating material ageing, such as that in a power cable. Thus it is hypothesized that by developing an EL model based on the factors contributing to the EL emission, the simulation results may provide some relevant information in understanding the ageing process thus allowing improved cable's lifetime estimation by monitoring the remaining cable's lifetime while in service. Therefore, it is necessary to develop an understanding of the underlying processes of EL and to investigate the possible influencing factors that contributed to EL emission before modelling techniques based on EL characteristics could be used reliably in service life of cable system.

1.2 Objectives

The main objectives of this research is to investigate the factors affecting the intensity of EL emission of polymeric insulating material as different condition produces different EL intensity by observing the occurrence of EL in LDPE at room temperature under the influence of uniform electric field configuration. The specific objectives of this research are listed as follows;

i. To investigate the EL intensity of virgin and aged LDPE samples under the application of high voltage stresses with variable frequency.

ii. To investigate the breakdown voltage and failure distribution of virgin and aged LDPE.

iii. To develop a mathematical model using Dimensional Analysis method that can be used for condition monitoring of insulating materials.

1.3 Scope of Work

In order to deliver the objectives of this thesis, the experimental data of EL measurements were conducted and collected at The Tony Davies High Voltage Laboratory, University of Southampton, United Kingdom. The EL experimental rig has been set up precisely in the laboratory for the experiments to be carried out. The EL setup utilizes a Peltier cooled electron multiplying charge coupled device (EMCCD) camera as its detection mechanism for measuring the light intensity. EMCCD camera is chosen over the traditional use of photomultiplier tube (PMT) because its fast framing, low light sensitivity, low-noise read out electronics, higher quantum efficiency and broader spectral range.

The study of EL phenomenon was undertaken on $100 \pm 5 \mu\text{m}$ virgin and aged additive-free LDPE. All materials under investigation were prepared at the clean

preparation room in the laboratory. Cleanliness is important throughout the sample preparation in order to eliminate discharges as much as possible. For aged LDPE, samples were thermally aged in fan oven either with varying elevated temperature (310 K, 330 K and 350 K) for 3 days or with varying duration (6, 12 and 21 days) at 330 K. To allow EL mechanism to take place, all materials must be coated with 20 nm thick gold layer which provides reasonable electrode conduction good optical transmission for the detecting EL phenomenon. Comparisons are made for EL emission between virgin and aged LDPE sample under the application of high electrical stresses in order to observe the effect of ageing processes towards EL emission.

To support the assumptions made on EL characteristics of LDPE, the optical, electrical and chemical properties of samples were analyzed through EL imaging, breakdown voltage, ultraviolet and visible (UV-Vis) spectrophotometer and Fourier Transform Infra-red (FTIR) spectroscopy which are conducted at The Tony Davies High Voltage Laboratory. The EL experiments were carried out as a function of varying applied AC stresses at a constant 50 Hz and of varying applied frequency at a constant applied voltage. The former was performed on virgin and aged LDPE while the latter was completed on virgin LDPE only. The EL measurements collected from the sample provide information on the point-on-wave measurement, phase angle as well as average EL intensity. All of these data are essential in deducing the EL characteristics of aged material.

With the growing importance of the use of EL phenomenon as diagnostic tool, it becomes necessary to develop a mathematical model that could replicate the experimental data thus allowing sound judgment in predicting the life of a cable. To achieve this, Dimensional Analysis method was used that incorporated the factors affecting the EL emission of both virgin and aged LDPE. The simulation data were compared with experimental data for validation of the model.

1.4 Significant Contributions

This thesis has established experimental methodology, procedures and sample preparation for the optical, electrical and chemical properties measurements for virgin and thermally aged LDPE. The UV-Vis spectroscopy, FTIR spectroscopy, gold sputtering technique, EL experimental rig and EMCCD camera were employed throughout the experiment. This study is concerned on the effect of ageing on the behavior of EL emission as a function of applied voltage as well as the effect of varying applied voltage and frequency on virgin LDPE. In general, the findings of this study are in agreement with other research outcomes from other researchers. However, there are several major contributions from this study as described below;

i. Statistical analysis was performed on the breakdown voltage distribution in virgin and aged LDPE using several statistical distributions such as Weibull, General Extreme Value and Johnson S_B based on Anderson-Darling and Kolgomorov-Smirnov goodness-of-fit. It has been known that Weibull distribution has been widely used in reliability engineering and life data analysis. However, it was found that Generalized Extreme Value provides the best-fitted distribution with lower statistical error, followed by Johnson S_B distribution in all breakdown analysis conducted on virgin and aged LDPE.

ii. EL emission measurements were conducted on LDPE samples that were thermally aged in fan oven either by varying the ageing temperature (at 310 K, 330 K and 350 K) for 3 days or by varying the ageing duration (at 6, 12 and 21 days) at 330 K. Several previous works have been done but were conducted separately thus comparisons between both types of thermally aged samples are impossible. This study allows comparisons to be made between both types of thermally aged samples in terms of optical and chemical properties as well as the EL emission characteristics hence contributing towards the knowledge of ageing processes in polymeric material. Both types produced different results in all measurements suggesting that the charge distributions in the polymer reacted differently depending on the method of ageing. This is also due to the oxidation process that was created as a result of thermal ageing (as observed from FTIR spectroscopy) thus modifying the chemical

properties of the aged samples.

iii. Investigations were also conducted on virgin LDPE as well to observe the effect of varying applied voltage at 50 Hz and varying applied frequency at a constant electric field on the light emission. The work on varying applied voltage concurred with other previous works by other researchers. However, the outcome of EL emission at varying applied frequency was the first of its kind. It was thought that the EL intensity will increase as the frequency is increased. Nonetheless, this was not materialized. The EL emission was irregular with increasing applied frequency but showed a similar pattern; the EL emission reached its peak at 20 Hz, 60 Hz and 90 Hz at all applied voltage. This suggests that the charge density is not a steady state but dependent on the recombination rate of the polymer. The findings of this research have contributed towards the advancement of theoretical knowledge related to the degradation processes in polymeric material.

iv. A mathematical model based on bipolar recombination model was developed using Dimensional Analysis method to simulate the EL phenomenon in LDPE to further understand the factors affecting the EL emission. The model is developed for the application of both virgin and aged LDPE. Many models have been constructed to simulate EL emission and many have reported the use of Dimensional Analysis in life model. Therefore, the mathematical model generated in this study is a novel work as none has been developed to simulate the EL emission using Dimensional Analysis method. Although the model developed is empirical at this stage, the information yielded from this model is valuable for future predictions. The outcome of this research may provide valuable information for high voltage cable engineers to predict the longevity of a cable before major catastrophic can occur. This could help the high voltage power company to save a lot of operational cost in reconstructing the high voltage system that might have been destroyed by the high voltage failure.

v. The results of this research can help to facilitate high voltage cable manufacturers in producing better cable insulating material with longer lifetime. Consequently, consumers will be able to enjoy better services and will have

minimum supply interruptions from high voltage cable breakdown. The findings from this study may help other researchers to gain deeper understanding and knowledge related to EL phenomenon and at the same time becomes a reliable reference for their research. This study wishes to inspire future researchers in improving the proposed method and model in order to further enhance the knowledge of EL phenomenon.

1.6 Outline of the Thesis

This thesis is divided into a few chapters and is arranged as follows:

Chapter 1 overviews the development of power cables for centuries, the space charges within the dielectric as one of the cause of degradation and the electroluminescence as a method to observe space charge distribution.

Chapter 2 discusses in details regarding the theories behind electroluminescence method and the factors affecting the EL intensity. Several previous works by various researchers are compared and discussed. This chapter also focuses on the background theory of Dimensional Analysis method as well as the mathematical steps to achieve dimensionally correct equation. This chapter also outlines the methods of model formulation and validation. The statistical analysis used in this work is also included in this chapter.

Chapter 3 describes extensively the experimental setup, the sample preparation and experimental procedures and measurements for this research. The results of the measurement are displayed and discussed in **Chapter 4**. The results include images taken from EL measurement, phase-resolved measurement for various factors and statistical analysis on the breakdown voltage of LDPE.

Chapter 5 focuses on the development of EL modelling using Dimensional Analysis method to simulate the phenomenon when LDPE is subjected to uniform AC field. The modelling is different for virgin and aged material. Each model is

formulated and validated through several methods. Finally, the conclusion and future works can be obtained in **Chapter 6**.

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