

New mathematical models for predicting the lifetime of EPDM insulators: Effect of elongation at break on the kinetic degradation of EPDM insulators subjected to thermo-oxidation

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ABSTRACT

Ethylene propylene diene monomer (EPDM) is an important polymer extensively exploited in platurgy. However, relatively few studies have been carried out to predict the lifetime of EPDM in different climatic conditions particularly, thermo-oxidation. Based on this realization, the aim of the present work was to develop mathematical models for predicting the lifetime of EPDM elastomers, used for insulation of electric cables.

The kinetic degradation of EPDM insulators, by monitoring change in a physical property (elongation at break test " ϵ_r "), was studied by following its thermo-oxidative aging (70, 90, 110 and 130 ° C in air circulating oven). The multiple linear regression analysis (MLRA), solved by the Cholesky method, was the mathematical approach developed in the modeling of the kinetic degradation. In this study, we used two insulators materials when the first insulator contained an amorphous EPDM and the second contained a semi-crystalline EPDM. The results showed that the polynomial models developed to predict elongation at break were reliable for both insulators under thermo-oxidation. The half-life times predicted by the mathematical models was found to be statistically significant ($p < 0.05$). In conclusion, the mathematical models developed in our study could be used confidently to predict the lifetime of EPDM elastomers.

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1. Introduction

The ethylene propylene diene monomer (EPDM) rubbers, obtained by polymerization, are synthetic copolymers of the elastomer family [1]. They are composed mainly of ethylene, propylene and a small proportion of diene. Due to their outstanding resistance to aging (thermal, atmospheric, radiative), EPDM elastomers are widely used in platurgy [2]. Indeed, they are used mainly for joints and insulation (car, roofing, cables, etc.). Today, EPDM production is growing exponentially.

EPDM elastomers are widely exploited in the industry and therefore it is considered expedient to estimate the in-service lifetimes of such rubbers [3-4]. The modelling of the kinetic degradation of EPDM under the influence of climatic factors (e.i. heat [5], UV radiation [6-8] and ozone [9]) was the subject of several studies to determine their lifetime. However, in contrast, there appears to be relatively little work carried out on the effects of thermal aging (thermo-oxidation) on the kinetic degradation of EPDM [10-12].

In view of the deficiencies concerning work carried out in past on degradation of EPDM elastomer, the objective of this work was to demonstrate the influence of thermal aging on the kinetic degradation of EPDM used for insulation of electric cables by predicting half-life times. Kinetic degradation was followed by monitoring changes in elongation-at-break as a function of exposure times. Mathematical kinetic models were employed in an attempt to simulate thermos-oxidative degradation.

2. Materials and experimental techniques

2.1. Materials

- The ethylene propylene diene (EPDM) elastomers used for preparation the two insulators were:
A semi-crystalline EPDM manufactured by Dow chemical Company under the trade name Nordel IP 3722P, composed of 70.5% by weight of ethylene, 29% of propylene and 0.5 % of 5-ethylidene-2-norbornene (ENB). Its density, specified by the manufacturer, is 0.88 g.cm and Mw =100 000 g/mol.
An amorphous EPDM manufactured by Dow chemical Company under the trade name Nordel IP 4520, composed of 50% by weight of ethylene, 45% of propylene and 5 % of 5-ethylidene-2-norbornene (ENB). Its density, specified by the manufacturer, is 0.86 g.cm Mw (g/mol) and the mass-average molar mass Mw =115 000 g/mol.
- Torada S22 (mineral oil) used as plasticizer and Flectol H or permanax TQ (trimethyldihydroquinoleine) and vulcanox (zincsalt of the mercaptobenzimidazole), as antioxidant agents,
- Aluminiumtrihydrate (ATH) as filler,
- Paraffin (white wax made up of saturated hydrocarbons), used as lubricant,
- Vinyl silane (beta methoxyethoxy), as coupling agent between fellers and elastomer,
- Perkadox vulcanization agent and TAC (triallyl-cianurate) cross-linking co-agent or activator,

2.2. Sample preparation

We prepared a square sheet at the National Company of Plastic and Rubber (ENPC) of Setif, Algeria. The mixing of the raw materials (elastomeric load, plasticizers and implementation agents) at 80 ° C, for half an hour was carried out using a cylinder mixer. After obtaining, the homogeneous mixture in the form of a sleeve and adding the crosslinks agent, the mixture was removed from the cylinder. The removed mixture was allowed to cool before proceeding to the vulcanization stage. Vulcanisation was carried out using compression-moulding techniques. Square sheets (200 × 200 × 2.0 mm) were moulded at 180°C using a cure time of 10 minutes and a compression force of 300 kN.

2.3. Mechanical tensile tests

The ϵ_r measurements were carried out using a universal traction machine of the "INSTRON MODEL 1185" type, according to the ASTM D882 standard. A strain-rate of 100 mm/minute was used. An average of five trials was performed to confirm the experimental results. Elongation at break was determined using the "equation (1)," [13-14]:

$$\epsilon_r = \left(\frac{L-L_0}{L_0} \right) \times 100 \quad (1)$$

Where:

- L_0 : Initial length of the specimen;
- L: Length of the specimen at break;
- ϵ_r : Percent elongation at break;

2.4. Accelerated aging (thermo-oxidation)

Dumbbell tensile test-pieces (type H₂), as shown in "Fig. 1," were used throughout the present study. Test-pieces were placed in circulating air-ovens; the temperatures used were 70, 90, 110 and 130°C. During the heating regime, test-pieces were removed at selected regular intervals and tested. Tests were carried out in triplicate.

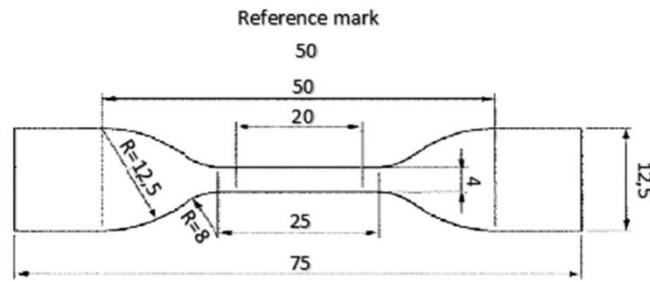


Figure 1. Dumbbell shape of tensile test pieces (type H₂)

2.5. Modelling

There are many ways for the modelling of physico-chemical phenomena among these methods: the general linear model (GLM) and the design of experiment [13], in our work, the best approach for the modelling of the kinetic degradation of EPDM insulators is the GLM. The most common general linear model used are derived from the general form given in “equation (2),” and “equation (3),” [14-15]:

$$\hat{Y} = \sum_{j=1}^m C_j \cdot f_j(X) \tag{2}$$

$$\hat{Y} = C_1 \cdot f_1(X) + C_2 \cdot f_2(X) + \dots + C_m \cdot f_m(X) \tag{3}$$

With:

Y: aging parameter;

X: exposure time;

C_j: Model Coefficients to be calculated (j = 1... m);

m: Number of coefficients;

f_j (X) : Regular Function, which may be in one of the following forms given in 4, 5, 6 and 7:

2.5.1. Monomial

$$f_j(X) = X^{j-1} \tag{4}$$

2.5.2. Exponential

$$f_j(X) = \alpha_j \cdot \exp (\beta_j \cdot X) \tag{5}$$

2.5.3. Logarithmic

$$f_j(X) = \alpha_j \cdot \sin(\beta_j \cdot X) + \gamma_j \cdot \cos (\delta_j \cdot X) \tag{6}$$

2.5.4. Trigonometric

$$f_j(X) = k_j \cdot \log (v_j \cdot X) \tag{7}$$

In this study, X is the exposure time and Y is the ϵ_r . We developed a model based on linear multiple regression analysis (LMRA) to predict ϵ_r . The linear systems resulting from LMRA were resolved using the Cholesky method. The modelling process consisted of changing the parameter m in the range [1, m_{max}] to obtain different linear models Y_m = (1, m_{max}). The best model (Y) with reasonable accuracy should satisfy four different statistical criteria defined as residual variance, coefficient of determination (r²_{max}), Student test and

Fisher–Snedecor test. We note that the confidence limit of the Student and Fisher–Snedecor tests is 95%, as supported by the following hypotheses: for Student test:

H_0 " $C_j = 0$ " against H_1 " $C_j \neq 0$ ", $j=1 \dots m$.

and for Fisher–Snedecor test:

H_2 " $C_1 = C_2 = C_3 \dots = C_m = 0$ "

If $F > t(n-m, \alpha/2)$, we reject the hypothesis H_0 , the model is then accepted.

If $F < t(n-m, \alpha/2)$ we accept H_0 , The model is then rejected.

With: $f(m-1, n-m; \alpha)$ is a value read from Fisher's table.

Our experimental results can be fitted well to a Gaussian [16].

2.6. Hardware and software used

To complete the work of modelling, we used a standard computer with a Intel (R) core (TM) i3-4160 CPU 3.60 GHz (4 CPUs) processor, a hard disk of 500 GB capacity and 4096 MB of RAM at the Laboratory of Applied Chemistry at the Centre for Biotechnology Research, Constantine. All our programs are written in MATLAB R2009b (64bit).

3. Results and discussions

The effect of thermo-oxidation at 70, 90, 110 and 130 ° C on the mechanical properties of EPDM insulators was evaluated by measuring the change in the percent ϵ_r as a function of the exposure time. The choice of ϵ_r is justified by its sensitivity to degradation. Conventionally, when the ϵ_r is a physical quantity widely used to measure the degradation and loss of 50% of the ϵ_r , which determines the half-life time of the material [14-15]. Conventionally, when the reaches 50% of that of the clean sample, the material is deemed out of service.

The change in the percent ϵ_r as a function of the exposure time for the semi-crystalline EPDM insulator and the amorphous EBDM insulator are shown in "Fig. 2," and "Fig. 3," respectively. Before aging, the elongation at break values of the semi-crystalline EPDM insulator and the amorphous EBDM insulator was 262 and 234 % respectively. This result is consistent with the recommendations of IEC 60502 standard, which requires a minimum value of 200% at 20 ° C.

After aging, from "Fig. 2," and "Fig. 3," it was found that the elongation began to increase from the initial value (before aging) to reach a maximum value and then a regression with a sudden drop was observed. This decrease is all the faster as the temperature of aging is greater (110 and 130 ° C) [17].

The increase in ϵ_r during the first exposure times has been attributed to the improvement in insulator quality due to crosslinking. The sudden drop in ϵ_r was explained by the chain scission reactions [18-20]. This phenomenon causes a decrease in the average molecular weight and in the degree of crosslinking on the one hand and in a loss of plasticizers on the other hand. Indeed, the chain scission process increases the mobility of the chains [21-22].

From the curve of "Fig. 2," and "Fig. 3," the half-life times of the two insulator was estimated at 9544, 3316, 1868 and 608 hours for semi-crystalline EPDM insulator and 5972, 2516, 1468 and 272 hours for the amorphous EPDM insulator exposed to 70, 90, 110 and 130 ° C respectively.

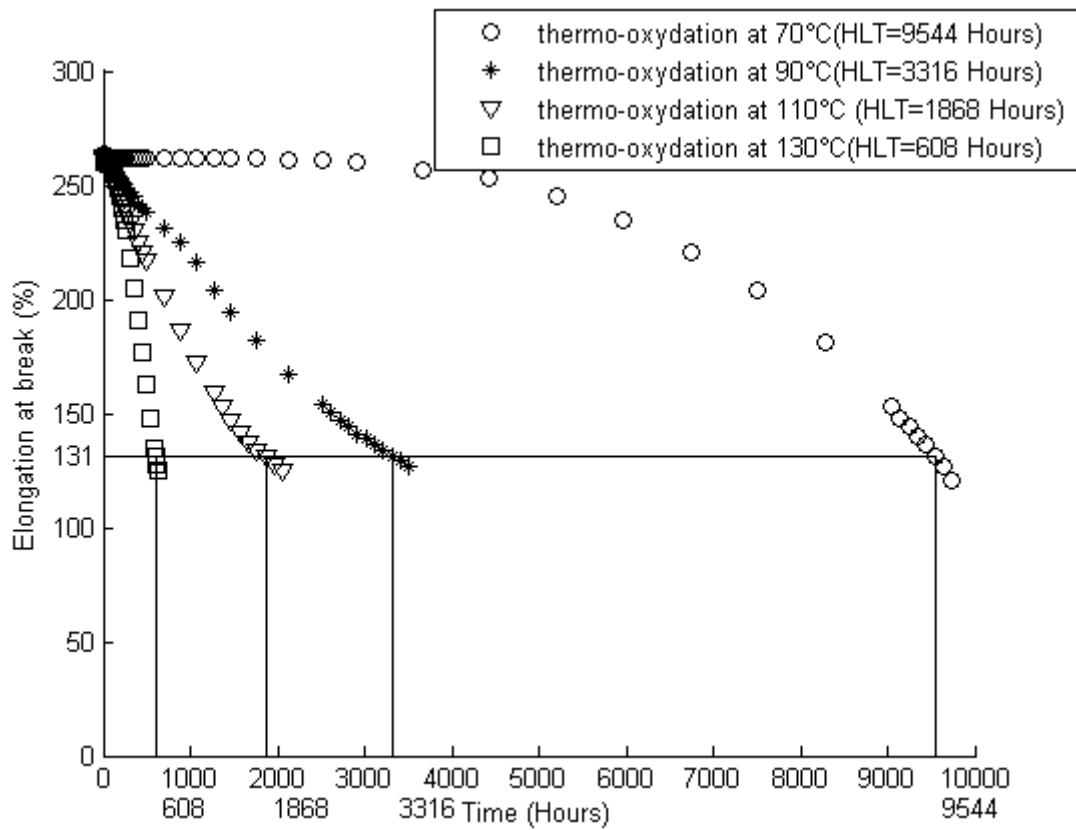


Figure 2. Elongation at break as a function of exposure time for semi-crystalline EPDM insulator, in thermo-oxidation at 70, 90, 110 and 130°C.

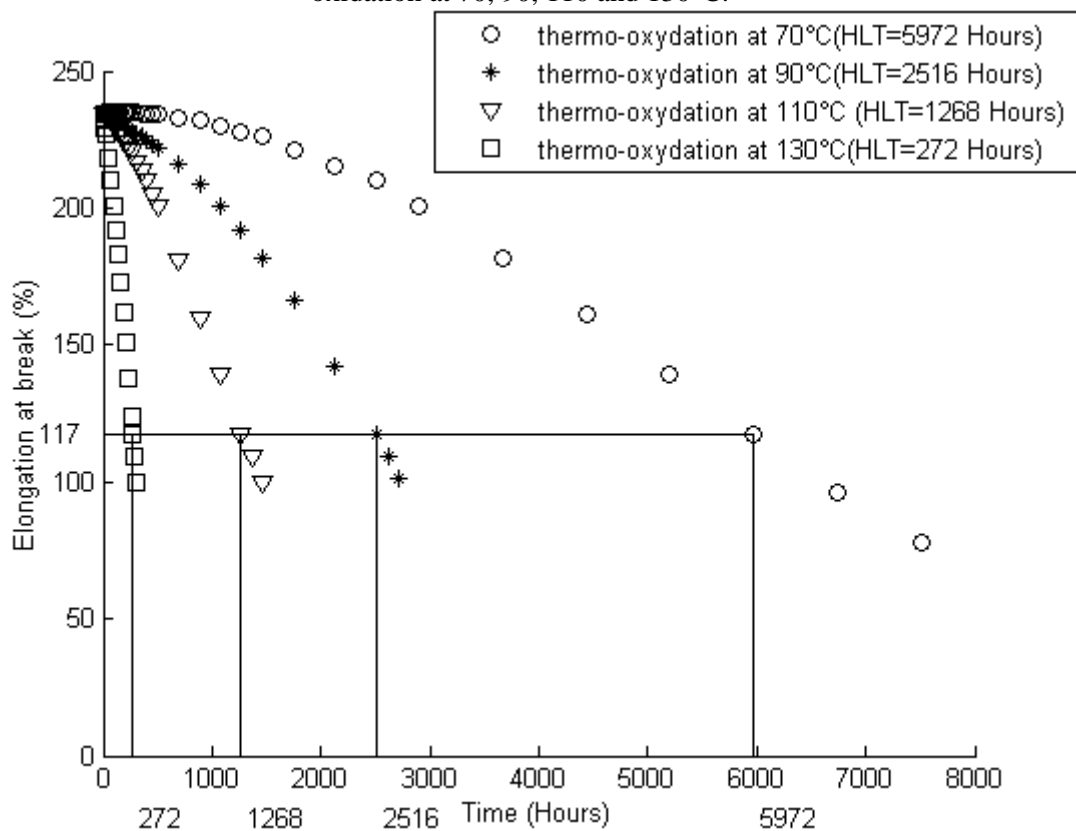


Figure 3. Elongation at break as a function of exposure time for amorphous EPDM insulator, in thermo-oxidation at 70, 90, 110 and 130°C.

4. Modelling

4.1. Modelling results

4.1.1. For the semi-crystalline EBDM insulator

In thermo-oxidation at 70 ° C

$$Y = 2,62 \cdot 10^2 - 88,74 \cdot 10^{-5} \cdot X + 62,25 \cdot 10^{-8} \cdot X^2 - 20,65 \cdot 10^{-11} \cdot X^3 \tag{8}$$

Table 1. Elongation at break for semi-crystalline EPDM insulator, in thermo-oxidation at 70°C.

N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values	N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values
1	0	262	262,35	-0,35	24	1076	262	261,86	0,14
2	2	263	262,35	0,65	25	1268	262	261,80	0,20
3	4	263	262,34	0,66	26	1460	262	261,74	0,26
4	6	264	262,34	1,66	27	1748	262	261,60	0,40
5	15	262	262,34	-0,34	28	2132	261	261,29	-0,29
6	20	262	262,33	-0,33	29	2516	261	260,77	0,23
7	44	262	262,31	-0,31	30	2900	260	259,98	0,02
8	68	262	262,29	-0,29	31	3668	257	257,28	-0,28
9	92	262	262,27	-0,27	32	4436	253	252,64	0,36
10	116	262	262,25	-0,25	33	5204	245	245,49	-0,49
11	140	262	262,24	-0,24	34	5972	235	235,28	-0,28
12	164	262	262,22	-0,22	35	6740	221	221,44	-0,44
13	188	262	262,20	-0,20	36	7508	204	203,40	0,60
14	212	262	262,19	-0,19	37	8276	181	180,62	0,38
15	236	262	262,17	-0,17	38	9044	153	152,52	0,48
16	260	262	262,16	-0,16	39	9144	148	148,44	-0,44
17	308	262	262,13	-0,13	40	9244	144	144,26	-0,26
18	356	262	262,10	-0,10	41	9344	140	139,98	0,02
19	404	262	262,08	-0,08	42	9444	136	135,59	0,41
20	452	262	262,06	-0,06	43	9544	131	131,11	-0,11
21	500	262	262,03	-0,03	44	9644	127	126,51	0,49
22	692	262	261,96	0,04	45	9744	121	121,81	-0,81
23	884	262	261,91	0,09					

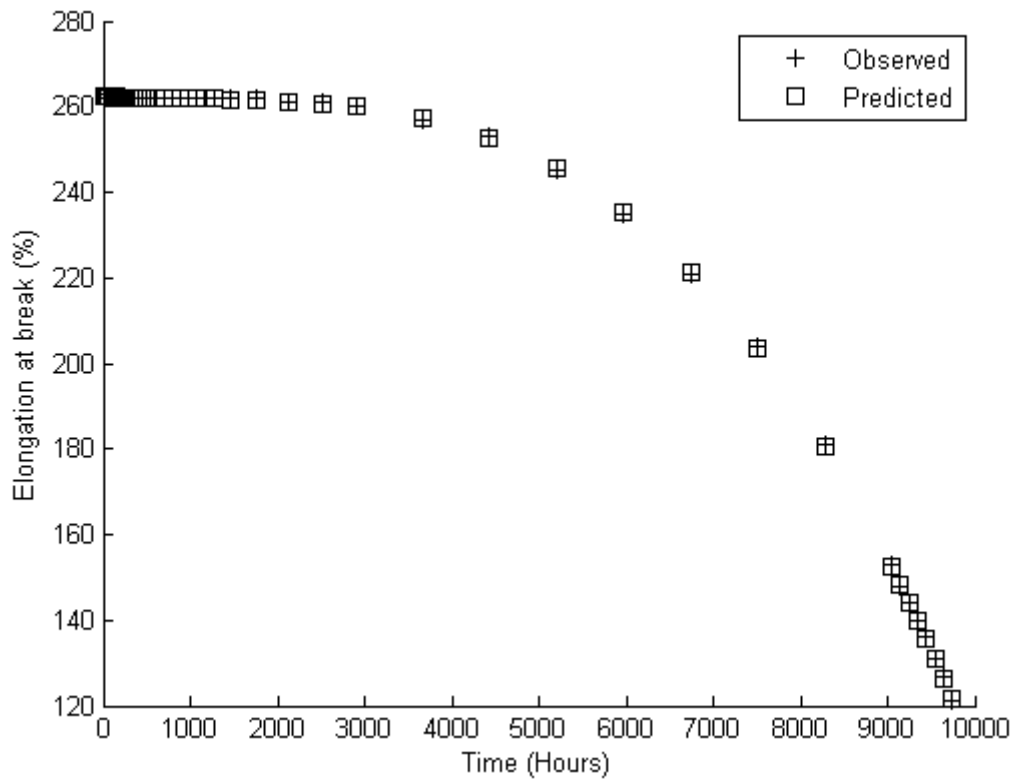


Figure 4. Comparison between the observed and predicted values of elongation at break for semi-crystalline EPDM insulator, in thermo-oxidation at 70 °C.

In thermo-oxidation at 90 ° C

$$Y = 2,61 \cdot 10^2 - 43,07 \cdot 10^{-3} \cdot X + 31,30 \cdot 10^{-7} \cdot X^2 + 12,96 \cdot 10^{-10} \cdot X^3 \quad (9)$$

Table 2. Elongation at break for semi-crystalline EPDM insulator, in thermo-oxidation at 90°C.

N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values	N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values
1	0	262	261,18	0,82	21	500	238	239,02	-1,02
2	2	262	261,09	0,91	22	692	231	230,30	0,70
3	4	261	261,01	-0,01	23	884	225	221,55	3,45
4	6	261	260,92	0,08	24	1076	216	212,82	3,18
5	15	261	260,53	0,47	25	1268	204	204,17	-0,17
6	20	261	260,32	0,68	26	1460	194	195,65	-1,65
7	44	260	259,28	0,72	27	1748	182	183,24	-1,24
8	68	258	258,24	-0,24	28	2132	167	167,68	-0,68
9	92	257	257,19	-0,19	29	2516	154	153,63	0,37
10	116	256	256,14	-0,14	30	2616	150	150,28	-0,28
11	140	255	255,09	-0,09	31	2716	147	147,06	-0,06
12	164	254	254,04	-0,04	32	2816	144	144,00	0,00
13	188	253	252,98	0,02	33	2916	141	141,09	-0,09
14	212	251	251,92	-0,92	34	3016	139	138,35	0,65
15	236	250	250,86	-0,86	35	3116	136	135,78	0,22
16	260	249	249,79	-0,79	36	3216	134	133,39	0,61
17	308	247	247,65	-0,65	37	3316	131	131,18	-0,18
18	356	245	245,51	-0,51	38	3416	129	129,17	-0,17
19	404	242	243,35	-1,35	39	3516	127	127,37	-0,37
20	452	240	241,19	-1,19					

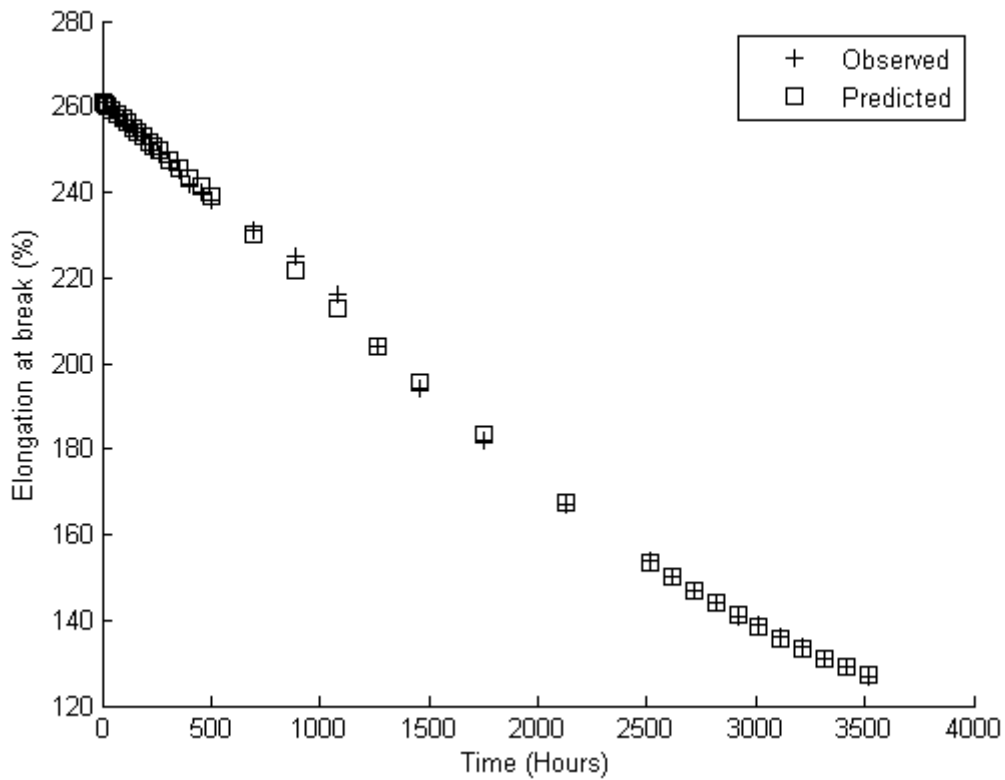


Figure 5. Comparison between the observed and predicted values of elongation at break for semi-crystalline EPDM insulator, in thermo-oxidation at 90 °C.

In thermo-oxidation at 110 °C

$$Y = 2,63 \cdot 10^2 - 95,90 \cdot 10^{-3} \cdot X + 55,91 \cdot 10^{-7} \cdot X^2 + 41,47 \cdot 10^{-10} \cdot X^3 \tag{10}$$

Table 3. Elongation at break for semi-crystalline EPDM insulator, in thermo-oxidation at 110°C.

N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values	N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values
1	0	262	263,04	-1,04	18	356	230	229,79	0,21
2	2	263	262,84	0,16	19	404	225	225,48	-0,48
3	4	263	262,65	0,35	20	452	221	221,21	-0,21
4	6	263	262,46	0,54	21	500	217	217,00	0,00
5	15	262	261,60	0,40	22	692	201	200,72	0,28
6	20	261	261,12	-0,12	23	884	186	185,49	0,51
7	44	259	258,83	0,17	24	1076	172	171,48	0,52
8	68	257	256,54	0,46	25	1268	159	158,87	0,13
9	92	254	254,26	-0,26	26	1368	153	152,92	0,08
10	116	252	251,99	0,01	27	1468	147	147,41	-0,41
11	140	250	249,73	0,27	28	1568	142	142,39	-0,39
12	164	247	247,48	-0,48	29	1668	137	137,87	-0,87
13	188	245	245,23	-0,23	30	1768	134	133,87	0,13
14	212	243	242,99	0,01	31	1868	131	130,42	0,58
15	236	241	240,77	0,23	32	1968	128	127,56	0,44
16	260	238	238,55	-0,55	33	2068	125	125,29	-0,29
17	308	234	234,15	-0,15					

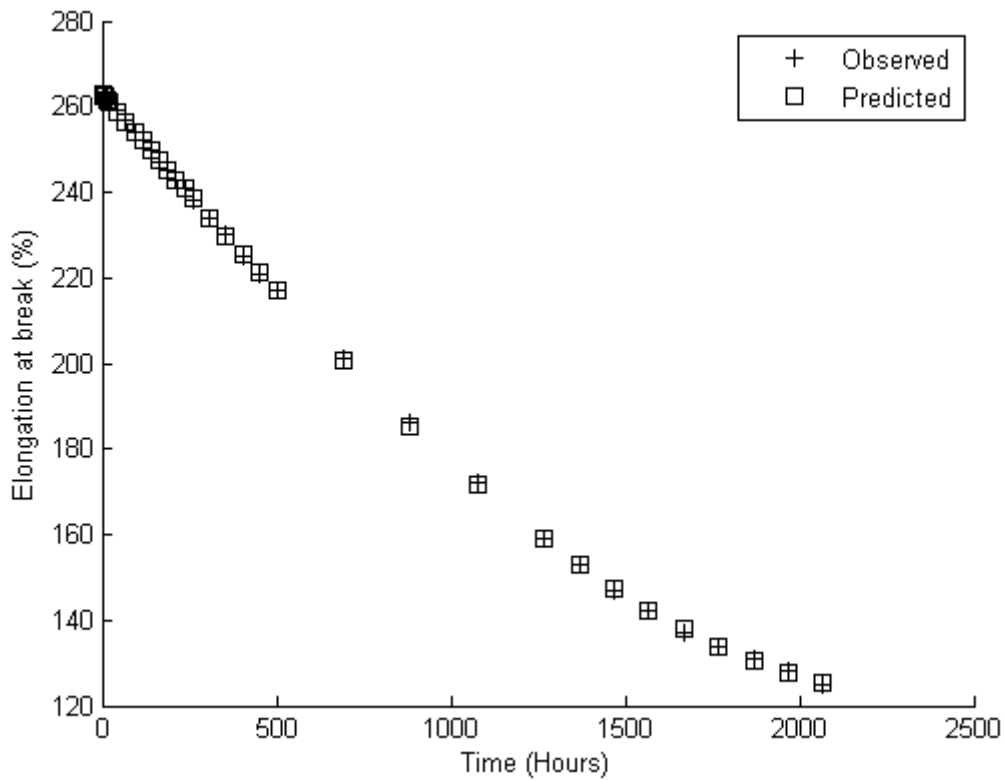


Figure 6. Comparison between the observed and predicted values of elongation at break for semi-crystalline EPDM insulator, in thermo-oxidation at 110 °C.

In thermo-oxidation at 130 °C

$$Y = 2,60 \cdot 10^2 + 30,01 \cdot 10^{-3} \cdot X - 69,34 \cdot 10^{-5} \cdot X^2 + 48,54 \cdot 10^{-8} \cdot X^3 \tag{11}$$

Table 4. Elongation at break for semi-crystalline EPDM insulator, in thermo-oxidation at 130°C.

N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values	N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values
1	0	262	260,31	1,69	14	212	240	240,13	-0,13
2	2	260	260,36	-0,36	15	236	235	235,15	-0,15
3	4	260	260,42	-0,42	16	260	230	229,77	0,23
4	6	260	260,46	-0,46	17	308	218	217,95	0,05
5	15	260	260,60	-0,60	18	356	205	205,01	-0,01
6	20	261	260,63	0,37	19	404	191	191,26	-0,26
7	44	260	260,33	-0,33	20	452	177	177,03	-0,03
8	68	259	259,29	-0,29	21	500	163	162,63	0,37
9	92	258	257,58	0,42	22	548	148	148,40	-0,40
10	116	255	255,22	-0,22	23	596	135	134,64	0,36
11	140	252	252,25	-0,25	24	608	131	131,32	-0,32
12	164	249	248,72	0,28	25	620	128	128,05	-0,05
13	188	245	244,67	0,33	26	632	125	124,84	0,16

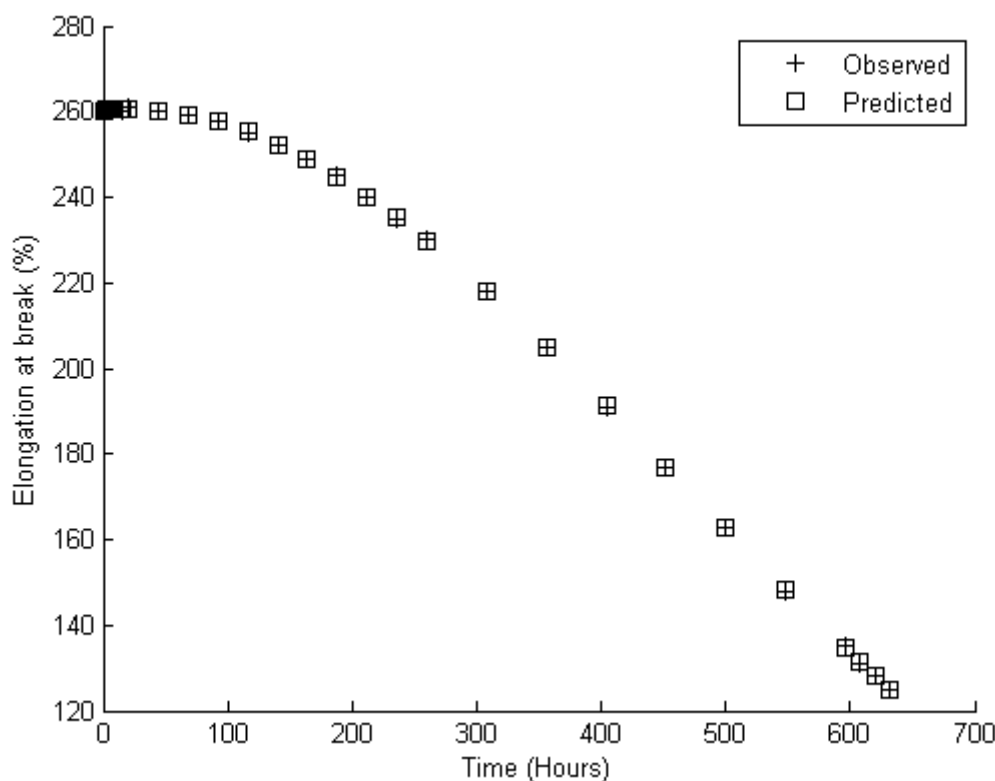


Figure 7. Comparison between the observed and predicted values of elongation at break for semi-crystalline EPDM insulator, in thermo-oxidation at 130 °C.

4.1.2. For the amorphous EBDM insulator

In thermo-oxidation at 70 °C

$$Y = 2,34 \cdot 10^2 + 66,93 \cdot 10^{-4} \cdot X - 89,64 \cdot 10^{-7} \cdot X^2 + 77,34 \cdot 10^{-11} \cdot X^3 \tag{12}$$

Table 5. Elongation at break for amorphous EPDM insulator, in thermo-oxidation at 70°C.

N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values	N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values
1	0	234	234,77	-0,77	19	404	234	234,50	-0,50
2	2	235	234,77	0,23	20	452	234	234,35	-0,35
3	4	235	234,77	0,23	21	500	234	234,18	-0,18
4	6	235	234,78	0,22	22	692	233	233,25	-0,25
5	15	235	234,79	0,21	23	884	232	231,95	0,05
6	20	235	234,80	0,20	24	1076	230	230,31	-0,31
7	44	235	234,83	0,17	25	1268	228	228,34	-0,34
8	68	235	234,85	0,15	26	1460	226	226,04	-0,04
9	92	235	234,86	0,14	27	1748	221	222,03	-1,03
10	116	235	234,87	0,13	28	2132	215	215,71	-0,71
11	140	235	234,88	0,12	29	2516	210	208,38	1,62
12	164	235	234,87	0,13	30	2900	201	200,17	0,83
13	188	235	234,86	0,14	31	3668	182	181,61	0,39
14	212	235	234,85	0,15	32	4436	161	160,98	0,02
15	236	235	234,83	0,17	33	5204	139	139,28	-0,28
16	260	235	234,80	0,20	34	5972	117	117,46	-0,46
17	308	235	234,72	0,28	35	6740	96	96,52	-0,52
18	356	234	234,62	-0,62	36	7508	78	77,42	0,58

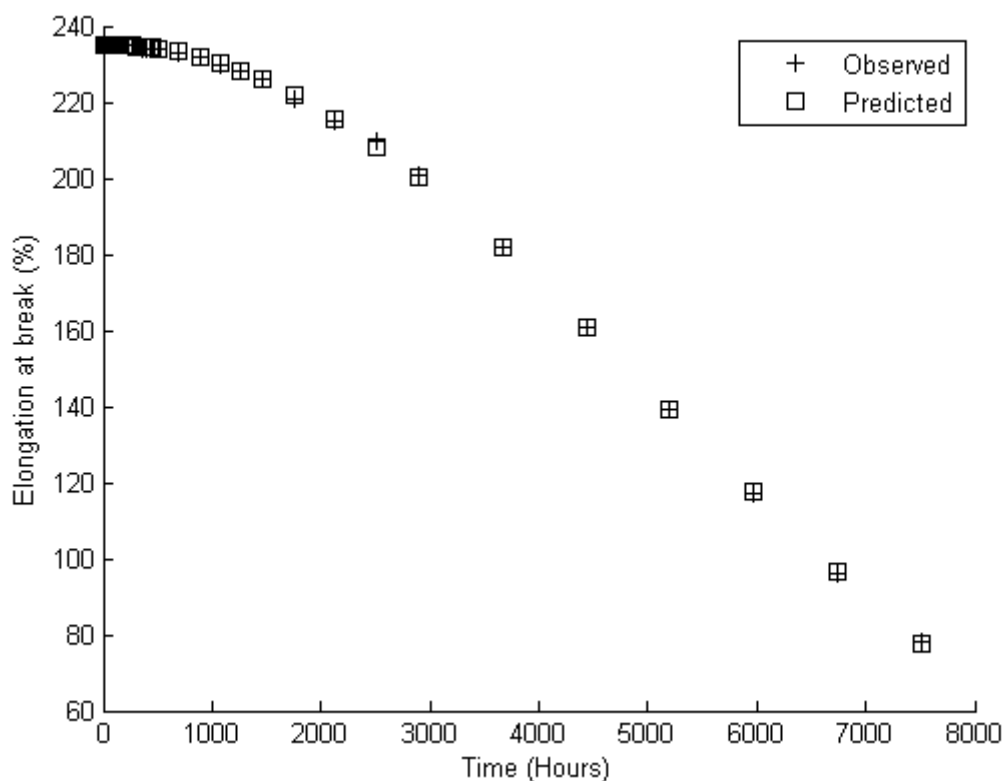


Figure 8. Comparison between the observed and predicted values of elongation at break for amorphous EPDM insulator, in thermo-oxidation at 70 °C.

In thermo-oxidation at 90 °C

$$Y = 2,34 \cdot 10^2 - 76,16 \cdot 10^{-3} \cdot X + 74,97 \cdot 10^{-7} \cdot X^2 + 14,73 \cdot 10^{-10} \cdot X^3 \tag{13}$$

Table 6. Elongation at break for amorphous EPDM insulator, in thermo-oxidation at 90°C.

N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values	N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values
1	0	234	233,91	0,09	17	308	227	227,36	-0,36
2	2	234	233,87	0,13	18	356	226	226,11	-0,11
3	4	234	233,84	0,16	19	404	225	224,81	0,19
4	6	234	233,80	0,20	20	452	223	223,45	-0,45
5	15	234	233,65	0,35	21	500	222	222,03	-0,03
6	20	233	233,56	-0,56	22	692	216	215,78	0,22
7	44	233	233,13	-0,13	23	884	209	208,64	0,36
8	68	233	232,68	0,32	24	1076	201	200,63	0,37
9	92	232	232,22	-0,22	25	1268	192	191,80	0,20
10	116	232	231,74	0,26	26	1460	182	182,16	-0,16
11	140	231	231,24	-0,24	27	1748	166	166,27	-0,27
12	164	231	230,73	0,27	28	2132	142	142,55	-0,55
13	188	230	230,21	-0,21	29	2516	117	116,17	0,83
14	212	230	229,67	0,33	30	2616	109	108,89	0,11
15	236	229	229,11	-0,11	31	2716	101	101,45	-0,45
16	260	228	228,54	-0,54					

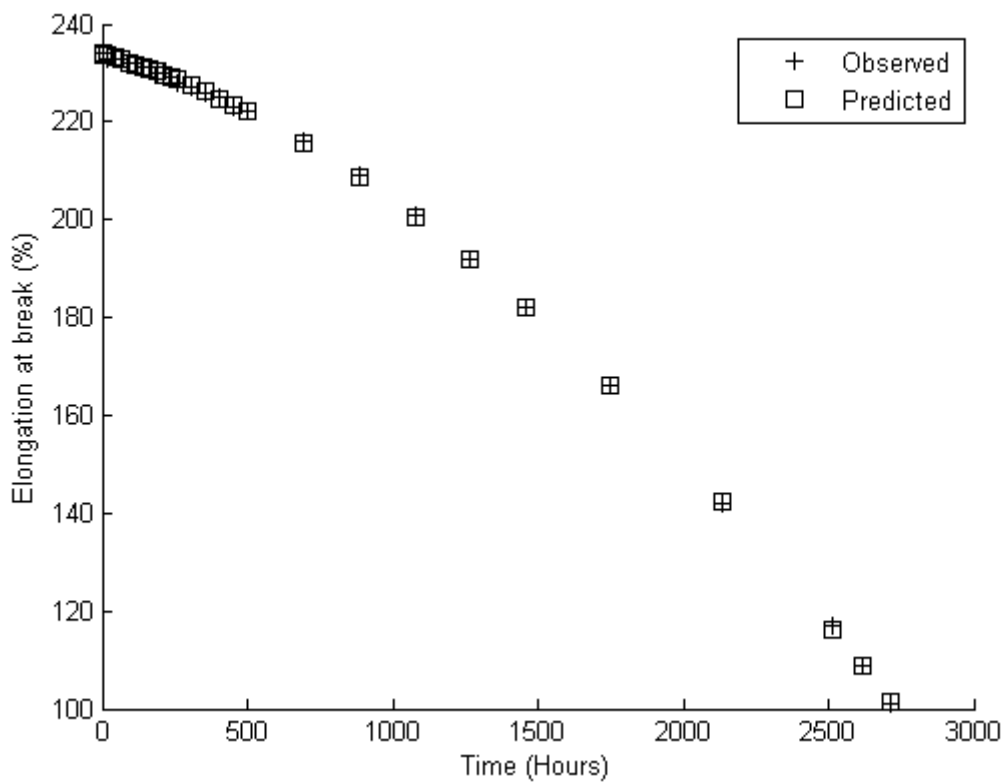


Figure 9. Comparison between the observed and predicted values of elongation at break for amorphous EPDM insulator, in thermo-oxidation at 90 °C.

In thermo-oxidation at 110 °C

$$Y = 2,34 \cdot 10^2 - 98,87 \cdot 10^{-3} \cdot X + 82,60 \cdot 10^{-7} \cdot X^2 + 35,27 \cdot 10^{-10} \cdot X^3 \quad (14)$$

Table 7. Elongation at break for amorphous EPDM insulator, in thermo-oxidation at 110°C.

N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values	N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values
1	0	234	234,27	-0,27	15	236	222	222,62	-0,62
2	2	234	234,21	-0,21	16	260	221	220,97	0,03
3	4	234	234,15	-0,15	17	308	217	217,44	-0,44
4	6	234	234,09	-0,09	18	356	214	213,65	0,35
5	15	234	233,81	0,19	19	404	210	209,59	0,41
6	20	234	233,64	0,36	20	452	205	205,30	-0,30
7	44	233	232,79	0,21	21	500	201	200,81	0,19
8	68	232	231,84	0,16	22	692	181	181,15	-0,15
9	92	231	230,79	0,21	23	884	160	159,88	0,12
10	116	230	229,65	0,35	24	1076	139	138,41	0,59
11	140	228	228,41	-0,41	25	1268	117	118,15	-1,15
12	164	227	227,09	-0,09	26	1368	109	108,55	0,45
13	188	226	225,68	0,32	27	1468	100	99,87	0,13
14	212	224	224,19	-0,19					

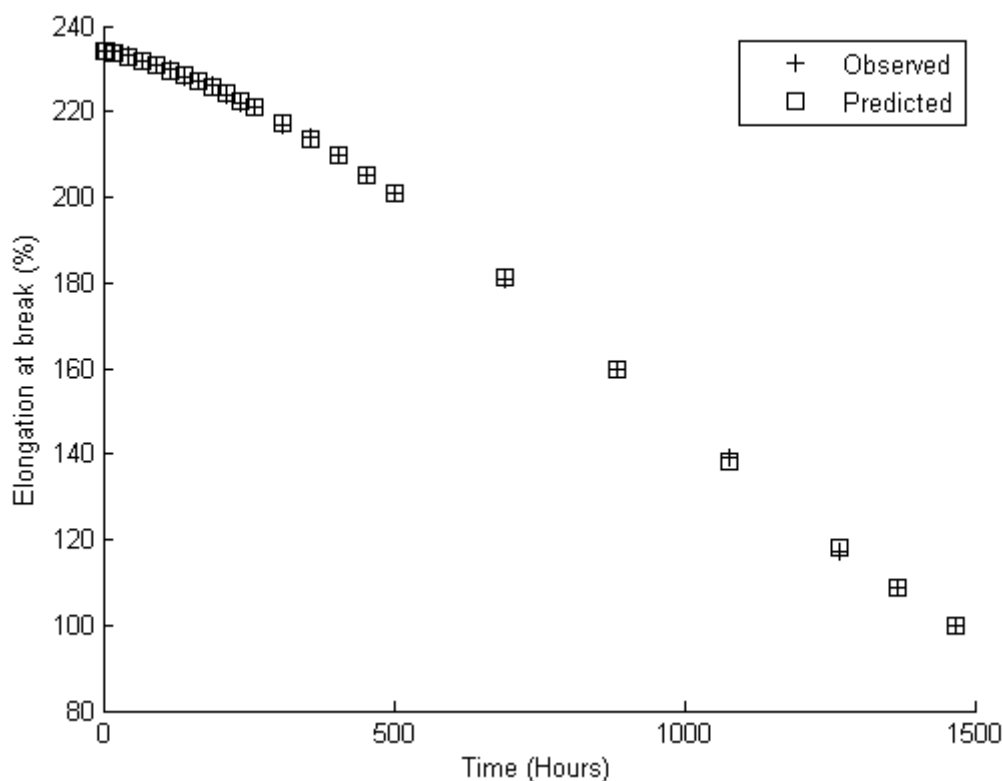


Figure 10. Comparison between the observed and predicted values of elongation at break for amorphous EPDM insulator, in thermo-oxidation at 110 °C.

In thermo-oxidation at 130 °C

$$Y = 2,34 \cdot 10^2 - 37,40 \cdot 10^{-2} \cdot X + 33,84 \cdot 10^{-5} \cdot X^2 - 20,32 \cdot 10^{-7} \cdot X^3 \tag{15}$$

Table 8. Elongation at break for amorphous EPDM insulator, in thermo-oxidation at 130°C.

N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values	N°	Time (Hours)	Observed (% ϵ_r)	Predicted (% ϵ_r)	Residual values
1	0	234	234,20	-0,20	11	140	183	182,90	0,10
2	2	233	233,46	-0,46	12	164	173	173,00	0,00
3	4	233	232,71	0,29	13	188	162	162,34	-0,34
4	6	232	231,97	0,03	14	212	151	150,76	0,24
5	15	229	228,66	0,34	15	236	138	138,07	-0,07
6	20	227	226,84	0,16	16	260	124	124,11	-0,11
7	44	218	218,23	-0,23	17	272	117	116,61	0,39
8	68	210	209,70	0,30	18	284	109	108,72	0,28
9	92	201	201,08	-0,08	19	296	100	100,44	-0,44
10	116	192	192,20	-0,20					

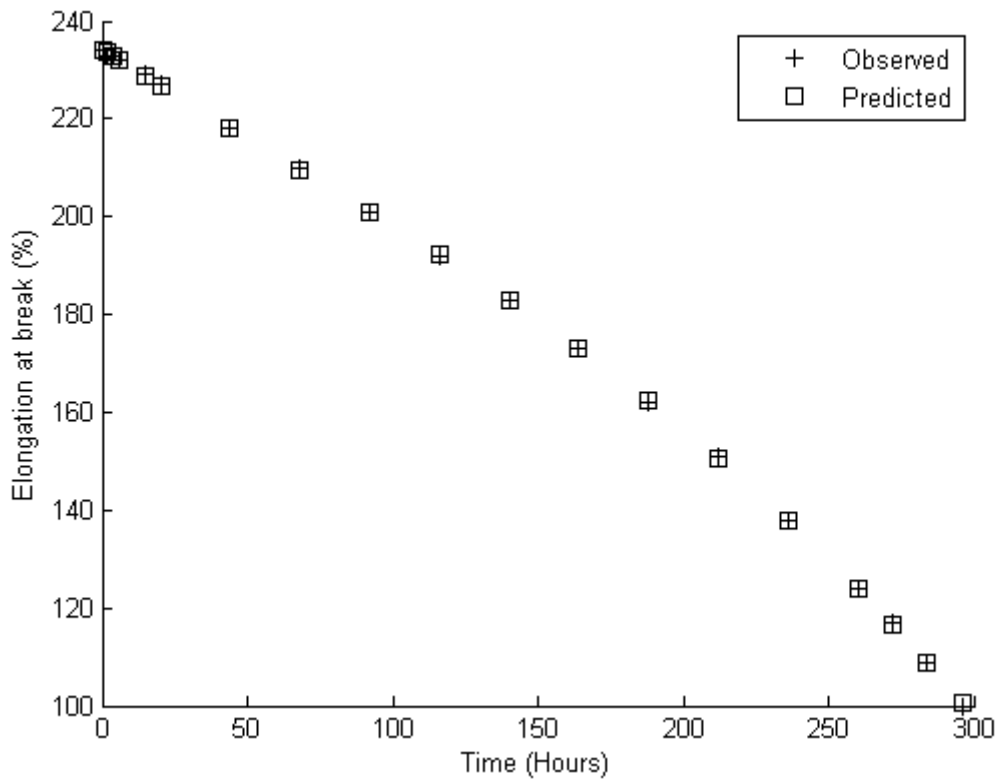


Figure 11. Comparison between the observed and predicted values of elongation at break for amorphous EPDM insulator, in thermo-oxidation at 130 °C.

Table 9. Statistical criteria of validity of the model obtained for semi-crystalline EPDM insulator (thermo-oxidation at 70, 90, 110 and 130 °C).

Aging by thermo-oxidation at 70 ° C				
Residual variance	Coefficient of determination (%)			Fisher-Snedcor test
0,19	99,99	Calculated value		Tabulated value
				$f(n-m,m-1,\alpha)$
				$1,10 \cdot 10^5$
				1,87
Student tests: $T(n-m, \alpha/2)=1,98$				
	T(C ₁)	T(C ₂)	T(C ₃)	T(C ₄)
	$2,43 \cdot 10^3$	4,75	11,68	54,35
Aging by thermo-oxidation at 90 ° C				
Residual variance	Coefficient of determination (%)			Fisher-Snedcor test
1,10	99,96	Calculated value		Tabulated value
				$f(n-m,m-1,\alpha)$
				$1,81 \cdot 10^4$
				1,87
Student tests: $T(n-m, \alpha/2)=1,98$				
	T(C ₁)	T(C ₂)	T(C ₃)	T(C ₄)
	$8,24 \cdot 10^2$	32,10	3,19	6,69
Aging by thermo-oxidation at 110 ° C				

Residual variance	Coefficient of determination (%)		Fisher-Snedcor test	
0,18	99,99	Calculated value	Tabulated value	
			$f(n-m,m-1,\alpha)$	
		$8,72 \cdot 10^4$	1,87	
	Student tests: $T(n-m, \alpha/2)=1,98$			
	T(C ₁)	T(C ₂)	T(C ₃)	T(C ₄)
	$1,79 \cdot 10^3$	$1,02 \cdot 10^2$	4,71	10,37
Aging by thermo-oxidation at 130 ° C				
Residual variance	Coefficient of determination (%)		Fisher-Snedcor test	
0,24	99,99	Calculated value	Tabulated value	
			$f(n-m,m-1,\alpha)$	
		$5,24 \cdot 10^4$	1,87	
	Student tests: $T(n-m, \alpha/2)=1,98$			
	T(C ₁)	T(C ₂)	T(C ₃)	T(C ₄)
	$1,27 \cdot 10^3$	8,01	43,84	28,16

Table 10. Statistical criteria of validity of the model obtained for amorphous EPDM insulator (thermo-oxidation at 70, 90, 110 and 130 °C).

Aging by thermo-oxidation at 70 ° C				
Residual variance	Coefficient of determination (%)		Fisher-Snedcor test	
0,25	99,98	Calculated value	Tabulated value	
			$f(n-m,m-1,\alpha)$	
		$4,88 \cdot 10^4$	1,87	
	Student tests: $T(n-m, \alpha/2)=1,98$			
	T(C ₁)	T(C ₂)	T(C ₃)	T(C ₄)
	$1,78 \cdot 10^3$	5,33	49,92	32,52
Aging by thermo-oxidation at 90 ° C				
Residual variance	Coefficient of determination (%)		Fisher-Snedcor test	
0,12	99,99	Calculated value	Tabulated value	
			$f(n-m,m-1,\alpha)$	
		$7,52 \cdot 10^4$	1,87	
	Student tests: $T(n-m, \alpha/2)=1,98$			
	T(C ₁)	T(C ₂)	T(C ₃)	T(C ₄)
	$2,01 \cdot 10^3$	27,05	20,65	4,14
Aging by thermo-oxidation at 110 ° C				
Residual variance	Coefficient of determination (%)		Fisher-Snedcor test	
0,16	99,99	Calculated value	Tabulated value	
			$f(n-m,m-1,\alpha)$	
		$5,48 \cdot 10^4$	1,87	

Student tests: T (n-m, $\alpha/2$)=1,98			
T(C ₁)	T(C ₂)	T(C ₃)	T(C ₄)
1,57. 10 ³	23,09	36,29	26,84
Aging by thermo-oxidation at 130 ° C			
Residual variance	Coefficient of determination (%)	Fisher-Snedcor test	
		Calculated value	Tabulated value
0,08	99,99	9,36. 10 ⁴	$f(n-m,m-1,\alpha)$ 1,87
Student tests: T (n-m, $\alpha/2$)=1,98			
T(C ₁)	T(C ₂)	T(C ₃)	T(C ₄)
1,66. 10 ³	61,84	6,33	16,64

- The residual variances tends to zero. This confirms that all the information about the model are appropriate.
- The values obtained of the coefficient of determination is closer to one, which confirms a good fit.
- The calculated values of Student's tests indicate that all the model coefficients are retained because they are higher than the tabulated value.

5. Estimated half-life time of EPDM by simulation

Table 11 presents the results of the half-life times for the EPDM insulators, obtained in thermo-oxidation. From the results recorded in Table 11, we observe good agreement between the observed values and the values predicted by the models. The degradation kinetics of EPDM insulators are influenced by the increase in the temperature [23, 24].

Table 11. Comparison between the observed and predicted values of the HLT for EPDM insulators.

		Thermo-oxidation			
		70°C	90°C	110 °C	130°C
Semi-crystalline EPDM Insulator	Observed values (Hours)	9544	3316	1868	608
	Predicted values (Hours)	9546,30	3325,13	1851,28	609,17
Amorphous EPDM Insulator	Observed values (Hours)	5972	2516	1268	272
	Predicted values (Hours)	5988,99	2503,91	1280,02	271,38

6. Conclusion

The results of our work showed that the polynomial models developed to predict the Half-life time of EPDM elastomers, used for insulation of electric cables under thermo-oxidation are reliable. These models are validated by various statistical criteria. Indeed, the results indicate a low residual variance, a coefficient of determination close to unity, and high values of Student coefficients and Fisher-Snedecor. The half-life time values predicted by the models are very close to those obtained experimentally. Finally, the polynomial models developed in our study can contribute to predict the half-life time of EPDM elastomers, used for insulation of electric cables. However, their applicability is likely valid only in an interval of time which depends on the exposure conditions. This requires knowledge of aging mechanisms and kinetics to develop a realistic lifetime model.

In perspective, we suggest using the design of experiment (DOE) to develop mathematical models to predict the life time of EPDM insulators, then compare the results obtained by the both (GLM and DOE) methods.

References

- [1] A. A. Athawale and A. M. Joshi, "Electronic Applications of Ethylene Propylene Diene Monomer Rubber and Its Composites," In *Flexible and Stretchable Electronic Composites*, D. Ponnamma, K. K. Sadasivuni, C. Wan, S. Thomas and M. A. AlMa'adeed, Eds, Springer International Publishing: New York, pp. 305–33. 2016.
- [2] A. N. Gent, "Engineering with rubber: how to design rubber components," 3 edition, Carl Hanser Verlag GmbH Co KG, 2012.
- [3] C. S. Woo and H. S. Park, "Useful lifetime prediction of rubber component. Engineering Failure Analysis," vol. 18, pp.1645-51, 2011.
- [4] C. S. Woo, S. S. Choi, S. B. Lee and H. S. Kim "Useful Lifetime Prediction of Rubber Components Using Accelerated Testing," *IEEE Transactions on Reliability*, vol. 59, pp. 11-7, 2010.
- [5] N. S. Tomer, F. Delor-Jestin, R. P. Singh and J. Lacoste, "Cross-linking assessment after accelerated ageing of ethylene propylene diene monomer rubber," *Polymer Degradation and Stability*, vol. 92, pp. 457-63, 2007.
- [6] Q. Zhao, X. Li and J. Gao, "Aging behavior and mechanism of ethylene-propylene-diene monomer (EPDM) rubber in fluorescent UV/condensation weathering environment," *Polymer Degradation and Stability*, vol. 94, pp. 339-43, 2009.
- [7] Q. Zhao, X. Li and J. Gao "Aging of ethylene-propylene-diene monomer (EPDM) in artificial weathering environment," *Polymer Degradation and Stability*, vol. 92, pp. 1841-1846, 2007.
- [8] S. D. Kucuk, H. Gerengi and Y. Guner "The Effect of Tinuvin Derivatives as an Ultraviolet (UV) Stabilizer on EPDM Rubber," *Periodicals of Engineering and Natural Sciences*, vol. 6, pp. 52-62, 2018
- [9] M. Giurginca, T. Zaharescu and A. Meghea, "Degradation of ethylene-propylene elastomers in the presence of ozone," *Polymer Degradation and Stability*, Vol. 50, pp. 45-8, 1995.
- [10] D. Bouguedad, A. Mekhaldi, A. Boubakeur and O. Jbara, "Thermal ageing effects on the properties of ethylene-propylene-diene monomer (EPDM) ," *Annales de chimie, Lavoisier*, vol. 33, pp. 303-313, 2008.
- [11] F. Delor-Jestin, J. Lacoste, N. Barros-Oudin, C. Cardinet and J. Lemaire, "Photo-, thermal and natural ageing of ethylene-propylene-diene monomer (EPDM) rubber used in automotive applications. Influence of carbon black, crosslinking and stabilizing agents," *Polymer Degradation and Stability*, vol. 67, pp. 469-477, 2000.
- [12] M. M. Abdel-Aziz and A. A. Basfar, "Aging of ethylene-propylene diene rubber (EPDM) vulcanized by γ -radiation," *Polymer Testing*, vol. 19, pp. 591-602, 2000.
- [13] B. Durakovic, "Design of Experiments Application, Concepts, Examples: State of the Art," *Periodical of Engineering and Natural Sciences*, Vol. 5, pp. 421-439, 2017.
- [14] M. Kaci, T. Sadoun, K. Moussaceb and N. Akroune, "Modeling of degradation of unstabilized and HALS-stabilized LDPE films under thermo-oxidation and natural weathering conditions," *Journal of Applied Polymer Science*, vol. 82, pp. 3284-3292, 2001.
- [15] K. Moussaceb, "Modeling of the Kinetics Degradation of Unstabilized and HALS-Stabilized LDPE Films Under Thermo-Oxidation and Natural Weathering Conditions," *Arabian Journal for Science and Engineering*, vol. 37, pp. 1327-1338, 2012.
- [16] M. Neuilly, "Modélisation et Estimation des Erreurs de Mesure," Editions Lavoisier Tec et Doc., Paris, 1993.
- [17] V. Langlois, L. Audouin, J. Verdu and M. Meyer, "Vieillessement thermique du Polyéthylène, Journées d'études des polymères," JEPO18, Groupe français d'Etudes et d'Applications des Polymères, 1990.
- [18] A. F. Younan, A. M. Choneim, A. A. A. Tawfik and K. N. Abd-El-Nour, "Electrical and physical properties of ethylene-propylene-diene monomer (EPDM) rubber loaded with semi-reinforcing furnace black," *Polymer Degradation and Stability*, vol. 49, pp. 215-22, 1995.

- [19] R. A. Assink, K. T. Gillen and B. Sanderson, "Monitoring the degradation of a thermally aged EPDM terpolymer by ^1H NMR relaxation measurements of solvent swelled samples," *Polymer*, vol. 43, pp. 1349-1355, 2002.
- [20] Y. Mecheri, A. Medjdoub, A. Boubakeur and M. Boumerzoug, "Influence du mode de vieillissement thermique sur les propriétés du polyéthylène réticulé chimiquement," *Annales de Chimie, Sciences Des Matériau, Lavoisier*, vol. 31, pp. 571-582, 2006.
- [21] N. C. Billingham and P. D. Calvert, "The physical chemistry of oxidation and stabilisation of polyolefins," *Development in Polymer Stabilization*, vol 3, pp. 139-190, 1980.
- [22] N. C. Billingham, D. C. Bott and A. S. Manke, "Application of thermal analysis methods to oxidation and stabilization of polymers," *Developments in Polymer Degradation*, vol. 3, pp. 63-100, 1981.
- [23] R. S. Rajeev, S. K. De, Anil K. Bhowmick and Baby John, "Studies on thermal degradation of short melamine fibre reinforced EPDM, maleated EPDM and nitrile rubber composites," *Polymer Degradation and Stability*, vol. 79, pp. 449-463, 2003.
- [24] P. Sutanto, F. L. Laksmna, F. Picchioni and L. P. B. M. Janssen, "Modeling on the kinetics of an EPDM devulcanization in an internal batch mixer using an amine as the devulcanizing agent," *Chemical Engineering Science*, vol. 61, pp. 6442-6453, 2006.