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Application of General Regression Neural Network in Forecasting the Effect of 6PPd Antioxidant and Gamma Irradiation on NBR Rubber Mechanical Properties

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ABSTRACT

Different NBR rubber compounds are designed using different concentration of 6PPD antioxidant and different gamma irradiation doses. In this study, an attempt has been made to predict the crosslinking density and mechanical properties of rubber compounds using the general regression neural network (GRNN) technique. The GRNN are trained using the experimental results first for five gamma doses as input and different number (2 or 4) of corresponding mechanical properties as the program output. Two cases are studied first for NBR rubber with 2.5 phr 6PPD antioxidant, Second for NBR rubber without 6PPD antioxidant. After training the GRNN, it is simulated to estimate the rubber mechanical properties for twenty one gamma doses (including the five measured values) as input. In the first case two conditions are carried. The outputs in the first condition are crosslinking density with hardness and tensile strength with elongation at break. While the outputs of the second condition are the four mechanical properties together. The mean absolute percentage error (MAPE) of the second condition in the first case is smaller than that of the first condition. For this reason in case of rubber without 6PPD the outputs are taken as the four mechanical properties. The simulation results takes 15 sec for estimating mechanical properties corresponding to twenty one gamma doses, while the experimental results took approximately between seven to thirty days for five gamma doses. GRNN provides excellent predictions with a high degree of correlation depending on increasing the number of mechanical properties used in the training process. Also it predicts mechanical properties for a large number of doses that cannot be measured experimentally in a very short time.

1. INTRODUCTION

All polymers suffer modifications at their properties when subjected to ionizing radiation (1) in which for very high doses, a complete loss of their mechanical strength is obtained. It is well known that the exposure of crosslinking type rubbers to radiation provides improved stability and mechanical properties (2). Since the use of rubber is widely used in the nuclear industry, it is important that we insure that the aging of these materials doesn’t have adverse effects on these systems (3). High-end radiation brings about a big problem when it comes to materials. The major affect is the crosslinking of the rubber which leads to the strengthening of these materials, but can cause embrittlement.

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When rubbers are irradiated, free radicals and ions are formed that can react to produce cross linked rubbers (4). At high irradiation doses, ozone attack on rubber compounds causes characteristic cracking perpendicular to the direction of applied stresses. This degradation is caused by reactions of ozone with the double bonds in the rubber molecules. At high irradiation doses the mechanical properties are adversely affected due to the degradation induced by increasing crosslinking.

In order to control the effects of rubber oxidation, antioxidant materials are added to NBR rubber. The addition of antioxidants to rubber plays a good role in protection of the rubber against high gamma radiation (5). Therefore, it is obvious that the major characteristics required for antioxidants properties are migration to the surface of a rubber and reactivity towards ozone. The mechanism of migration to the surface and reaction with ozone and thereby keeping the rubber unattacked. N-(1, 3-dimethylbutyl)-N'-phenylphenylenediamine (6PPD) are still the most widely used antioxidants in rubber (6).

GRNN, as proposed by Donald F. Specht in (7) falls into the category of probabilistic neural networks. This neural network like other probabilistic neural networks needs only a fraction of the training samples. The data available from measurements of an operating system is generally never enough for a back propagation neural network (8&9). Therefore the use of a probabilistic neural network is especially advantageous due to its ability to converge to the underlying function of the data with only few training samples available (9&10). Therefore GRNN is a powerful tool to perform predictions and comparisons of system performance.

A study for the effect of 6PPD concentration on physical and mechanical properties of gamma irradiation crosslinked NBR is done in (11). This is very important from the view point of finding out optimum concentration of 6PPD required to obtain desired properties. This will help avoid the unnecessary addition of 6PPD in NBR which will in turn reduce the cost.

Specific goals of this paper are as follows: First, test the efficacy of GRNN for predicting mechanical properties of NBR rubber compounds with different concentration of 6PPD antioxidant and gamma irradiation. Second, comparing the GRNN model performance to an experimental results taken from (11) for five gamma doses (0, 500, 1000, 1500, 2000) KGy. Finally, predict the mechanical properties of rubber at different gamma irradiation doses located between the five gamma doses measured in the laboratory.

2. MARTIALS AND METHODS

2.1 Experimental

NBR rubber samples with different concentration of 6PPD antioxidant (1, 1.5, 2, 2.5 and 3) phr, were irradiated at gamma doses (0, 500, 1000, 1500, 2000) KGy (11). The mixing of the rubber is carried out on a laboratory two-roll mill (Farrel-UK, 152 mm & 330 mm) at a friction ratio of 1:1.4. The samples were vulcanized in a hydraulic press (Farrel-UK) at 153C$^{\circ}$ and pressure of 150 kg/cm^2 for a period of 5 minutes for NBR compounds.

2.1.1 Irradiation of Samples

The molded nitrile rubber samples were irradiated in air at room temperature by ^{60}Co source of gamma facility Canadian Gamma Chamber, and represented at the National Center for Radiation Research and Technology (NCRRT) with rate 2.7 KGy/hr is used for γ-irradiation. The irradiation times for the samples are shown in table 1.

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Table 1: The irradiation time for the samples

Dose [KGy]	Day	Hour
500	7	17
1000	15	10
1500	23	2
2000	30	20

2.1.2 Crosslinking Measurements

The crosslinking, ν_e, of the NBR rubber compounds was calculated by the equation:

$$\nu_e = \rho_r N / M_C \quad (1)$$

Where ρ_r: the density of the rubber

N: Avogadro's number = $6.02214179 \times 10^{23}$ mol^{-1}

M_c: the average molecular weight between crosslinks of rubber and can be calculated according to the

theory of Flory and Rehner(12) by:

$$M_C = - V_1 \rho_r \{ \Phi_r^{1/3} - \Phi_r / 2 \} / \{\ln (1 - \Phi_r) + \Phi_r + \mu \Phi_r^2 \} \quad (2)$$

Where V_1: the molar volume for toluene = 106.3 cm3/mole

Φ_r : the volume fraction of polymer and can be determined from the equilibrium degree of swelling $\mathbf{Q_m}$ as follows:

$$Q_m = 1/\Phi_r = 1 + \{M_S \rho_r / M_r \rho_S\} \quad (3)$$

Where M_r and M_S are the weights of dried rubber and absorbed solvent, respectively, also, ρ_s and ρ_r are the densities of the solvent used and the rubber compound, respectively.

μ is the Huggins (13) interaction parameter between solvent and polymer and it can be calculated as:

$$\mu = (\delta_s - \delta_r)^2 V_1 / R T \quad (4)$$

Where δ_s and δ_r are the solubility parameters of the solvent and rubber compounds respectively.
R: the Universal gas constant= 8.314 J·K^{-1}·mol^{-1}
T: the absolute temperature.

2.1.3 Mechanical Properties

The tensile strength and elongation at break (Eb) are measured by using a Zwick (Germany) Tensile Testing Machine (Model Z010) and a crosshead speed of 500mm/min using five dumb-bell tensile specimens being shaped according to ASTM D-412(14). The hardness test is measured by a Zwick (Germany) Hardness Tester Machine (Model 3150) according to ASTM D-2240(15).

In this study, two rubber compounds were selected with different 6PPD antioxidant concentration (0, 2.5) phr. Two variables of compound formulations have been considered in this analysis: gamma dose and 6PPD antioxidant concentration. Four output variables have been taken into account: crosslinking density, tensile strength, elongation at break and hardness as shown in table 2 & 3 respectively.

2.2 General Regression Neural Network (GRNN)

Generalized Regression Neural Network (GRNN) is a kind of Radial Basis Function (RBF) neural network with a one pass learning algorithm and highly parallel structure.

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GRNN was introduced by Specht in 1991[16] as a memory-based network that provides estimates of continuous variables. The algorithm provides smooth approximation of a target function even with sparse data in a multidimensional space. The advantages of GRNN are fast learning and easy tuning. The GRNN is composed of four layers: input, pattern (radial basis layer), summation and output as shown in figure 1.
Each neuron of the pattern layer uses a radial basis function as an activation function. This function is commonly taken to be Gaussian as follows:

$$G_j(\mathbf{x}) = \exp\left(-\frac{\left\|\mathbf{x}-\mathbf{C}_j\right\|^2}{s_j^2}\right) \qquad (5)$$

Where $\mathbf{C}_j$ is a center vector, s_j is a smoothing parameter or bandwidth and ||.|| is the Euclidean norm.

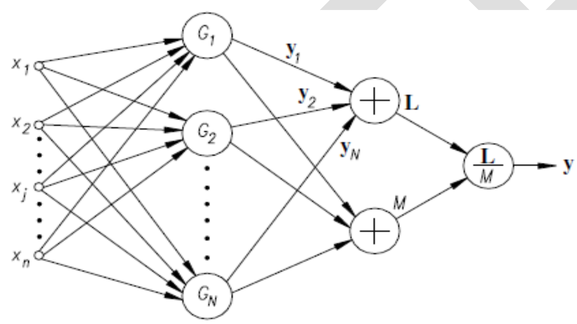

Fig. 1: GRNN Architecture

Each training vector is represented by one pattern neuron with the center $\mathbf{C}_j = \mathbf{x}_j$, $j = 1, 2, \ldots, N$. Where N is a number of training points. The neuron output expresses the similarity between the input vector $\mathbf{x}$ and the j-th training vector. So the pattern layer maps the n-dimensional input space into N-dimensional space of similarity. The GRNN output is an average of training y-patterns weighted by the degree of similarity between paired with them x-patterns and the query pattern:

$$\mathbf{y} = g(\mathbf{x}) = \frac{\sum_{j=1}^{N} G_j(\mathbf{x})\mathbf{y}_j}{\sum_{j=1}^{N} G_j(\mathbf{x})} \qquad (6)$$

Note that the GRNN generates a vector as an output. The dimension of this vector does not affect the number of parameters to estimate unlike in other popular models such as multilayer perceptron or neuro-fuzzy networks. This should be considered as a valuable property.
The performance of GRNN is related with bandwidths s_j governing the smoothness of the regression function. Determining optimal bandwidth values is a major problem in GRNN training.
The forecasting model similar to GRNN called Nadaraya-Watson estimator was presented in [17]. In this estimator the product kernel is used as RBF. The product kernel has different bandwidths for each component of $\mathbf{x}$. But for the different training patterns the same set of bandwidths are used. In the case

of GRNN with Gaussian functions for each training pattern there is only one bandwidth but for each pattern the bandwidth is different.

3. RESULTS AND DISCUSSION

The data of crosslinking density and mechanical properties of rubber compounds NBR with 2.5 phr 6PPD and NBR without 6PPD are obtained from [11], which are shown in tables 2 and 3 respectively. The GRNN are trained using the experimental results shown in tables 2 and 3. Five gamma doses are taken as input values and there four corresponding physical and mechanical properties as the program output for two cases (NBR with 2.5 phr 6PPD and NBR without 6PPD).

Table 2: Experimental values of NBR with 2.5 phr 6PPD antioxidant

Samples	Dose [KGy]	Antioxidants [phr]	Crosslinking Density $n_e x10^{+23}$	Tensile Strength [MPa]	Elongation at Break [%]	Hardness
1	0	2.5	0.1065	18.89	138.62	67.34
2	500	2.5	0.1926	16.93	60.21	73
3	1000	2.5	0.2651	15.91	33.89	80.66
4	1500	2.5	0.3091	17	31.48	79.22
5	2000	2.5	0.4462	18.81	26.7	83.48

Table 3: Experimental values of NBR without 6PPD antioxidant

Samples	Dose [KGy]	Antioxidants [phr]	Crosslinking Density $n_e x10^{+23}$	Tensile Strength [MPa]	Elongation at Break [%]	Hardness
1	0	0	0.0673	19.54	136.83	70.3
2	500	0	0.1783	19.58	44.74	83.62
3	1000	0	0.3368	9.51	13.44	79.74
4	1500	0	0.4154	5.52	0.01	92.56
5	2000	0	0.8549	6.31	0.01	94.43

3.1 Case 1: NBR Rubber with 2.5 phr 6PPD Antioxidant

In this case the five doses for 2.5phr 6PPD antioxidant are considered as GRNN input and its corresponding mechanical properties shown in table 2 as the output values for the training process. The GRNN is tested firstly to predict two outputs only, crosslinking with hardness as shown in table 4 and figure 2, and tensile strength with elongation as shown in table 5 and figure 3. Secondly, the GRNN is tested to predict the four mechanical outputs together and the results are illustrated in table 6 and figure 4.

3.1.1 Condition 1

In this condition the GRNN is trained by the experimental data given in table 2. First for five doses as the inputs and their corresponding hardness and crosslinking density as outputs, then the program is asked to estimate the crosslinking density and hardness values for 21 doses including the five experimental values. The GRNN results are shown in table 4. The error between GRNN prediction and experimental values, also the mean absolute percentage error (MAPE) for the five doses given in table 2 are calculated in table 4.

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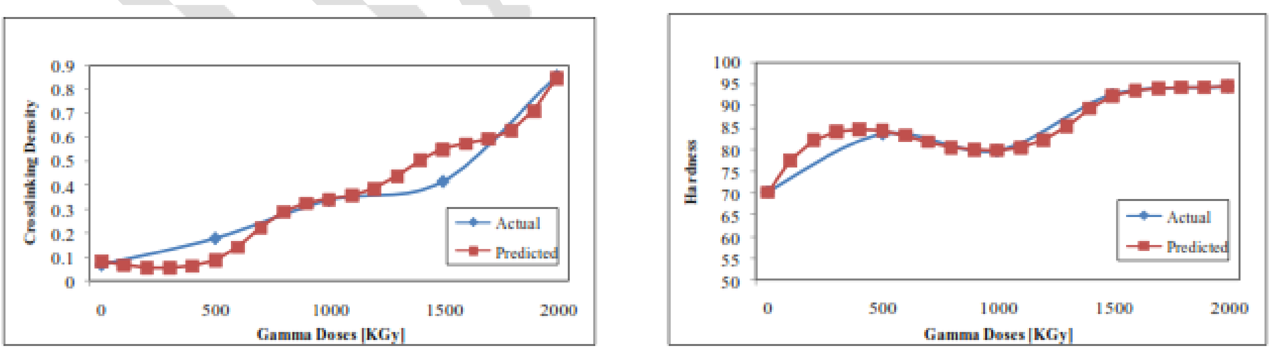
Table 4: Prediction of Crosslinking Density and Hardness for NBR with 2.5 phr 6PPD antioxidant

Input Dose [KGy]	Crosslinking Density $n_e x10^{+23}$	Crosslinking Error	Hardness	Hardness Error
0	0.1799	0.0733	69.2126	1.8726
100	0.2125		69.5999	
200	0.2633		70.2131	
300	0.3178		70.9004	
400	0.3565		71.4826	
500	0.3763	0.1837	72.0117	0.9882
600	0.3835		72.7344	
700	0.3832		73.9699	
800	0.3779		75.9182	
900	0.3702		78.3042	
1000	0.3630	0.0978	80.4179	2.42E-01
1100	0.3582		81.8116	
1200	0.3556		82.5600	
1300	0.3544		82.9173	
1400	0.3538		83.0763	
1500	0.3537	0.0445	83.1405	3.9205
1600	0.3537		83.1559	
1700	0.3540		83.1354	
1800	0.3546		83.0706	
1900	0.3558		82.9357	
2000	0.3579	0.0883	82.7016	0.7783
MAPE [%]		47.0911		2.0632

Figure 2, shows a comparison between the experimental values of (a) crosslinking density and (b) hardness (one input and two outputs) for five doses, and there predicted values for the twenty one doses.



(a) (b)

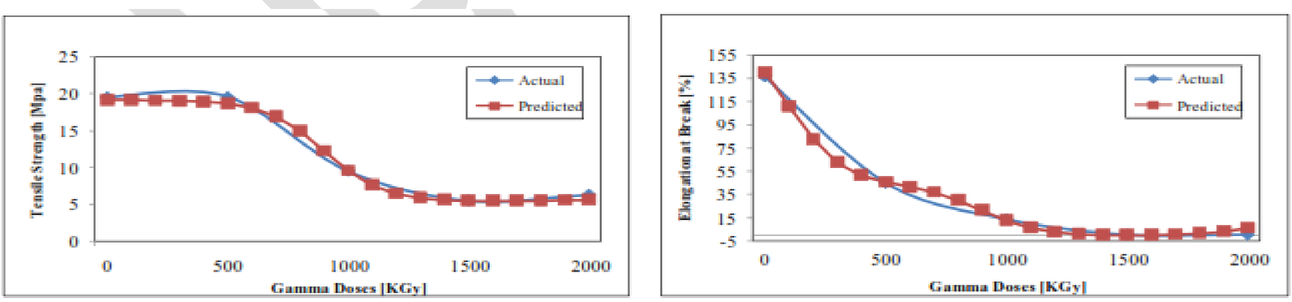
Fig .2: Comparison between the Five Experimental Values and Twenty One Predicted Values in Condition 1 for (a) Crosslinking Density and (b) Hardness

Table 5, shows the predicted values for tensile strength and elongation at break corresponding to twenty one doses as output values. Also the error between GRNN prediction and experimental values for the five doses are illustrated.

Table 5: Prediction of Tensile Strength and Elongation for NBR with 2.5 phr 6PPD antioxidant

Input Dose [KGy]	Tensile Strength [MPa]	Error in Tensile Strength	Elongation at Break [%]	Error in Elongation
0	18.8894	5.47E-04	138.8897	2.70E-01
100	18.7237		135.6195	
200	18.3098		127.4563	
300	17.4487		110.4528	
400	16.1873		85.3864	
500	15.0601	1.87	62.3762	2.17
600	14.4719		48.6716	
700	14.3773		42.1908	
800	14.6499		38.9079	
900	15.1975		36.5788	
1000	15.8473	0.0626	34.6020	0.7120
1100	16.3819		33.1346	
1200	16.7135		32.2568	
1300	16.8850		31.8105	
1400	16.9647		31.6063	
1500	16.9981	0.0018	31.5245	0.0445
1600	17.0075		31.5099	
1700	17.0001		31.5491	
1800	16.9750		31.6529	
1900	16.9303		31.8317	
2000	16.8742	1.94	32.0548	5.35
MAPE [%]		4.35		5.2181

Figure 3, shows a comparison between the experimental values of (a) tensile strength and (b) elongation at break for five doses and its predicted values for the twenty one doses including the five training values between them.



(a) (b)

Fig. 3: Comparison between Actual Values of Rubber Properties and Predicted Values by the GRNN in Condition 1 for (a) Tensile strength and (b) Elongation at Break

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3.1.2 Condition 2

In this condition the GRNN is trained with the experimental data for five doses as the input and its corresponding crosslinking density, tensile strength, elongation at break, and hardness as the output (one input and four outputs). Then the program is asked to estimate their values for 21 doses including the five experimental values. The GRNN results and the error between the actual and predicted values are shown in table 6.

Table 6: Prediction of the four mechanical properties for NBR with 2.5 phr 6PPD antioxidant

Input Dose [KGy]	Crosslinking Density $n_e x10^{+23}$	Error in Crosslinking	Tensile Strength [MPa]	Error in Tensile Strength	Elongation at Break [%]	Error in Elongation	Hardness	Error in Hardness
0	0.1065	4.83E-06	18.8899	2.82E-05	138.6228	2.81E-03	67.3390	9.37E-04
100	0.1248		18.6847		128.1787		68.4557	
200	0.1517		18.3840		112.8786		70.0917	
300	0.1850		18.0104		93.8690		72.1244	
400	0.2189		17.6309		74.5495		74.1904	
500	0.2471	0.0545	17.3165	0.3865	58.5360	1.6739	75.9033	2.9033
600	0.2667		17.0976		47.3572		77.1001	
700	0.2789		16.9632		40.4550		77.8407	
800	0.2860		16.8879		36.5091		78.2670	
900	0.2901		16.8493		34.3484		78.5057	
1000	0.2926	2.74E-02	16.8331	9.23E-01	33.1866	7.03E-01	78.6428	2.02
1100	0.2944		16.8318		32.5571		78.7319	
1200	0.2963		16.8433		32.1967		78.8072	
1300	0.2988		16.8700		31.9555		78.8948	
1400	0.3028		16.9182		31.7407		79.0210	
1500	0.3091	2.36E-06	17.0000	1.53E-05	31.4812	1.21E-03	79.2198	1.82E-04
1600	0.3194		17.1345		31.1062		79.5393	
1700	0.3357		17.3498		30.5322		80.0467	
1800	0.3608		17.6812		29.6613		80.8260	
1900	0.3973		18.1633		28.4004		81.9588	
2000	0.4463	1.03E-05	18.8101	0.0001	26.711	0.0115	83.4782	0.0017
MAPE [%]		7.7310		1.6172		0.9809		1.2963

Figure 4, shows a comparison between the experimental values of (a) crosslinking density, (b) tensile strength, (c) elongation at break and (d) hardness for five doses and its predicted values for the twenty one doses including the five training values between them. The results show that the experimental values for either of the crosslinking, tensile strength, elongation and hardness are similar to experimental.

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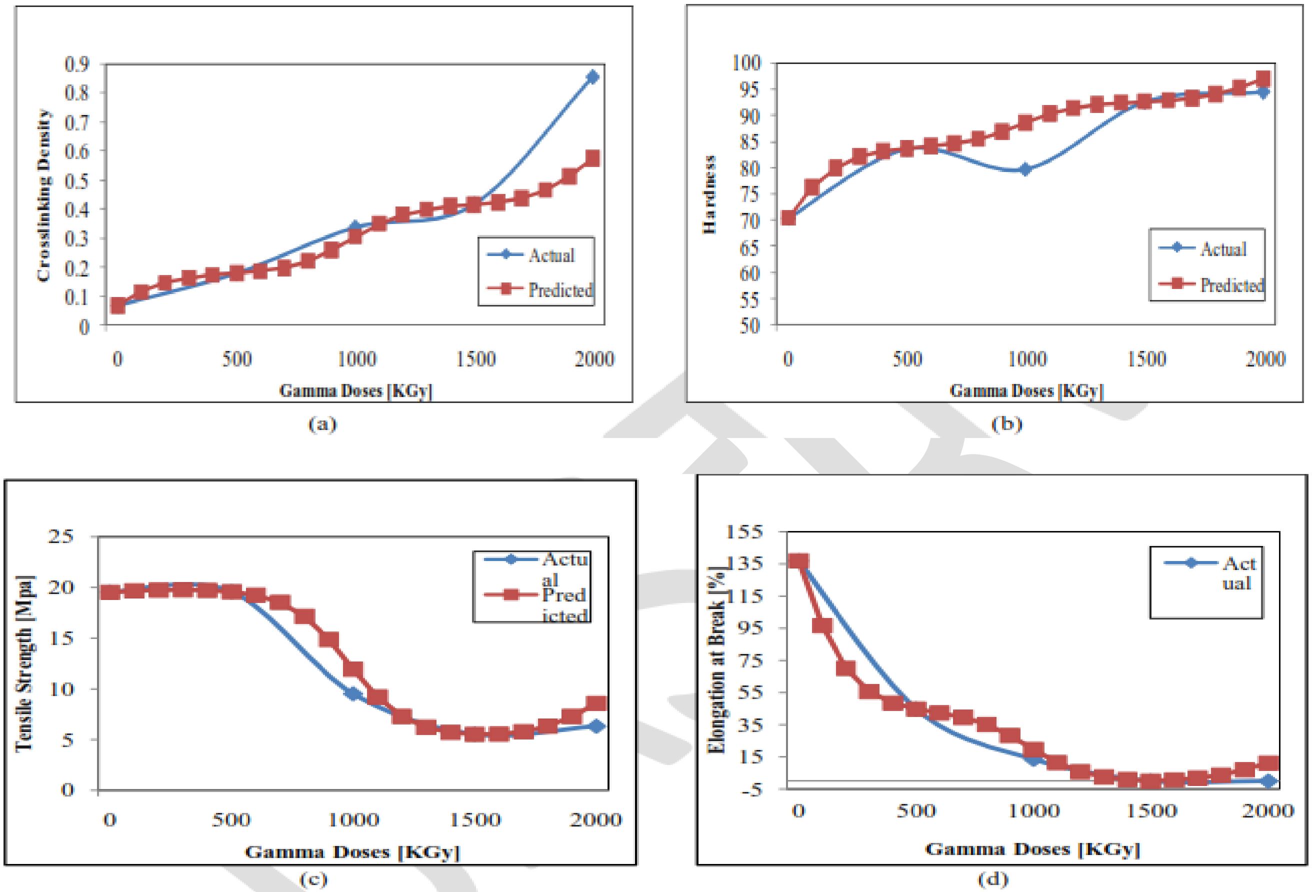

Fig. 4: Comparison between Actual Values of Rubber Properties and Predicted Values by the GRNN in Case of NBR Rubber with 2.5phr 6PPD Antioxidant (a) Crosslinking Density, (b) Hardness, (c) Tensile Strength and (d) Elongation at Break

The results show that the error between experimental values GRNN prediction for either of the crosslinking, tensile strength, elongation and hardness are smaller than that of condition 1. By comparing the MAPE in tables 4 and 5 with that in table 6, it is found that as the number of the output mechanical properties increase the MAPE decreases. For this reason condition 2 will be considered as the main approach for the predicting the mechanical properties of NBR rubber without 6PPD antioxidant.

3.2 Case 2: NBR Rubber without 6PPD Antioxidant

In this condition the GRNN is trained with the experimental data for five doses as the input and its corresponding crosslinking density, tensile strength, elongation at break, and hardness as the output (one input and four outputs). Then the program is asked to estimate their values for 21 doses including the five experimental values. The GRNN prediction, the error between the actual and predicted values and the MAPE are shown in table 7.

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Table 7: Prediction of the four mechanical properties for NBR without 6PPD antioxidant

Input Dose [KGy]	Crosslinking Density $n_e x10^{+23}$	Error Crosslinking	Tensile Strength [MPa]	Error Tensile Strength	Elongation at Break [%]	Error Elongation	Hardness	Error Hardness
0	0.0673	1.74E-05	19.5398	0.0001	136.8273	0.0026	70.2995	0.0004
100	0.1144		19.6715		96.5661		76.1041	
200	0.1455		19.7486		70.0729		79.9253	
300	0.1628		19.7674		55.6214		82.0136	
400	0.1721		19.7227		48.4223		83.0630	
500	0.1783	3.34E-06	19.5799	6.55E-06	44.7399	9.38E-05	83.6199	1.53E-05
600	0.1855		19.2464		42.2889		84.0307	
700	0.1978		18.5342		39.5354		84.5491	
800	0.2205		17.1514		35.1670		85.4139	
900	0.2576		14.8681		28.3028		86.7916	
1000	0.3051	0.0316	11.9396	2.4296	19.6037	6.1637	88.5437	8.8037
1100	0.3496		9.1945		11.4698		90.1844	
1200	0.3808		7.2836		5.7936		91.3318	
1300	0.3988		6.2177		2.5771		91.9873	
1400	0.4086		5.7097		0.9199		92.3376	
1500	0.4154	6.20E-08	5.5199	4.23E-06	8.38E-03	1.6250E-03	9.26E+01	1.17E-05
1600	0.4235		5.5388		0.7555		92.7953	
1700	0.4379		5.7722		1.8274		93.1918	
1800	0.4655		6.3169		3.7462		93.9399	
1900	0.5124		7.2786		6.9540		95.2060	
2000	0.5747	0.2802	8.5671	2.2571	0.00934	6.6000E-04	96.8859	2.4559
MAPE [%]		8.4438		12.2639		13.7427		2.7284

Figure 5, shows a comparison between the experimental values of crosslinking density, tensile strength, elongation and hardness for five doses and its predicted values for the twenty one doses including the five training values between them.

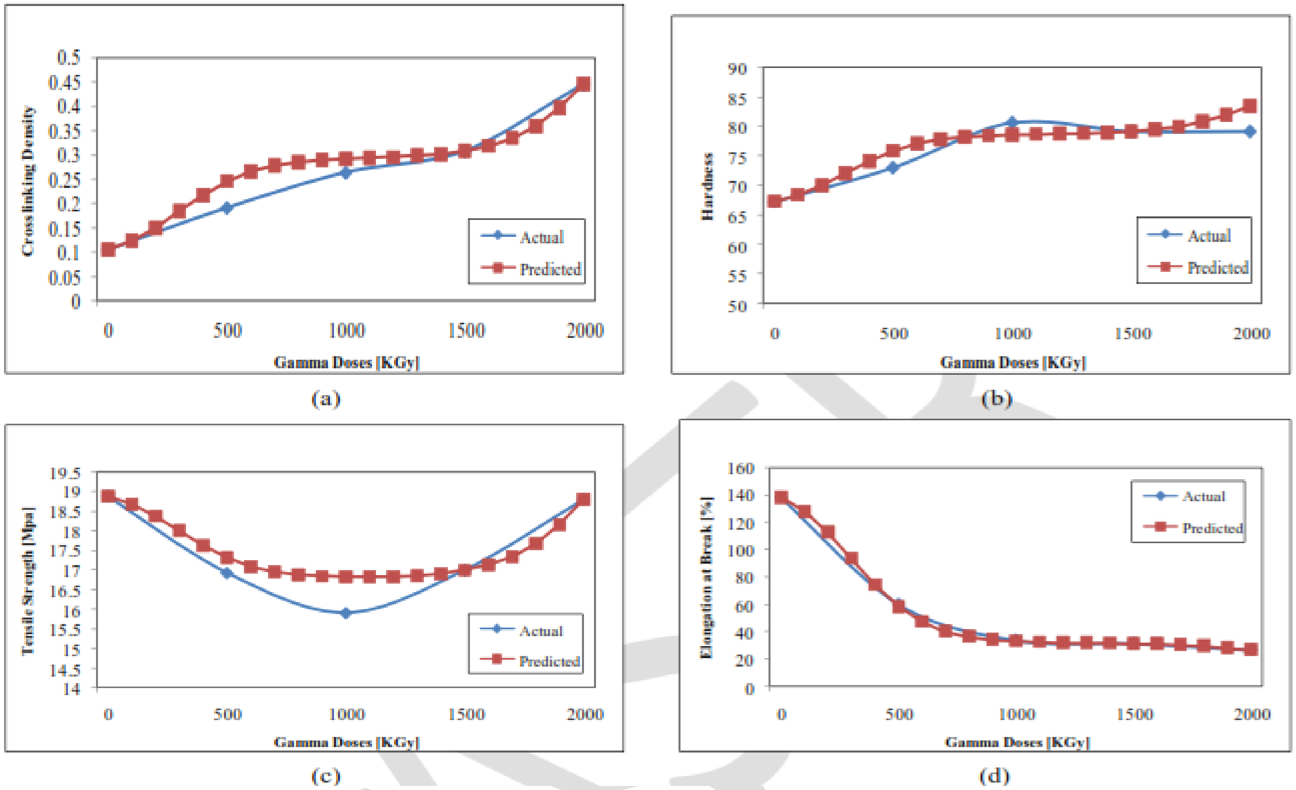

Fig. 5: Comparison between Actual Values of Rubber Properties and Predicted Values by the GRNN for (a) Crosslinking Density, (b) Hardness, (c) Tensile Strength and (d) Elongation at Break

Finally from the result of the two cases it is found that the mechanical properties corresponding to four doses experimentally take from 7 to 30 days for calculation as shown in table 1. While predicting these properties for 21 doses using GRNN takes 15 sec. The result indicates that prediction using GRNN is a powerful tool.

4. CONCLUSIONS

From the previous study, it is concluded that:

1. GRNN is implemented for predicting different mechanical properties (crosslinking density, tensile strength, elongation at break, and hardness of rubber) of NBR rubber, influenced by two significant factors (radiation dose and antioxidant concentration).
2. The predicted results done by the GRNN are almost very close to what is determined from the experimental results.
3. GRNN is a powerful and simple alternative technique for the prediction of rubber properties. It predicts the mechanical properties for a large number of doses (21 doses) in 15 sec that cannot be measured experimentally in this short time, as 5 doses measured in the laboratory took from 7 to 30 days.
4. It is found that the MAPE of prediction decreases with increasing the number of mechanical properties.

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