



## Investigation of the Factors Affecting Nonlinear Optical Properties by Statistical Analysis and Gray Relational Analysis Method

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Accepted 16 November 2022

### Abstract

In this study, it was aimed to statistically examine the hyperpolarization changes according to some reactivity indexes such as HOMO and LUMO energy, band gap energy, electronegativity, chemical hardness, softness and electronegativity index and to determine the most suitable molecule to be used in optoelectronic device applications. In the statistical analysis using the data obtained from the DFT study, it was determined that there was a significant and negative relationship between hyperpolarizability and LUMO energies, absolute electronegativity, energy gap and chemical hardness, and a positive relationship with softness and electronegativity. However, no significant relationship was found between HOMO energies and hyperpolarizability. In the analysis made by Gray Relational Analysis (GRA) method, it was determined that the most suitable molecule to be used in optoelectronic device applications was Naphthalene-2-carboxylic acid [2-hydroxy-2-(4-nitro-phenyl)-ethyl]-amide. It is thought that this study will guide the design of important and effective organic NLO molecules and will be of interest to researchers.

**Keywords:** Nonlinear optical materials, DFT analysis, Statistical analysis, Gray relational analysis

### 1. Introduction

Nonlinear optical (NLO) materials, which have been studied extensively in recent years, can be defined as materials in which light waves can interact with each other. The interest in these materials increased especially with the discovery of the laser in the 1960s and significant developments were experienced [1]. NLO materials have extensive applications in optoelectronic devices such as telecommunications, information storage, optical switching, signal processing [2-7]. Many experimental and theoretical studies have been carried out in the last twenty years on the synthesis and analysis of the properties of these materials, which have many important application areas [8-14]. On the other hand, DFT calculations have been extensively studied on the analysis of hyperpolarization ( $\beta$ ) properties of molecules that may show potential NLO properties [15, 16]. When these articles are examined, it is necessary to calculate the NLO properties correctly and to use extended basis sets. In particular, these basic sets should include the d and p polarization functions [17,18].

The design of efficient organic materials for applications in nonlinear optical effects relies on

asymmetric polarization caused by electron donating and electron accepting groups added to the molecule at appropriate positions in molecular systems. The location of the added groups is also vital. For this reason, it is important to examine the effect of placing different groups in different positions on the optical properties. In a study conducted by A. Kaya in 2019, the change in optical properties was investigated by adding groups with different electronegative properties to organic molecules [19]. In addition, hyperpolarization ( $\beta$ ) changes were investigated according to some reactivity indexes such as HOMO and LUMO energy, band gap energy ( $\Delta E$ ), electronegativity ( $\chi$ ), chemical hardness ( $\eta$ ), softness (S) and electronegativity index ( $\omega$ ) of molecules. In this study, the effect of different input parameters created by adding different groups on the NLO feature was investigated statistically.

In addition, it is aimed to determine which of the molecules may be suitable for use in optoelectronic device applications by Gray Relational Analysis (GRA) method, which is one of the multi-criteria decision-making methods.

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
The following sections of the study are organized as follows; In the second section, the material and method are explained, in the third section, the findings obtained from the study are given, and in the fourth section, the results and evaluations are made.

## 2. Materials and Methods

### 2.1. DFT Studies

In the study conducted by Kaya [19], 15 molecules containing different groups were studied. The molecules used in the study are summarised in Table 1.

Table 1. Molecules used in the study

 R'	R''	-H	-C <sub>6</sub> H <sub>5</sub>	- C <sub>6</sub> H <sub>4</sub> NO <sub>2</sub>	-C <sub>6</sub> H <sub>4</sub> Cl	-C <sub>6</sub> H <sub>4</sub> Br
-C <sub>10</sub> H <sub>7</sub>		1	2	3	4	5
-C <sub>6</sub> H <sub>4</sub> Cl		6	7	8	9	10
-C <sub>6</sub> H <sub>4</sub> Br		11	12	13	14	15

- 1- Naphthalene-2-carboxylic acid (2-hydroxy-ethyl)-amide,
- 2- Naphthalene-2-carboxylic acid (2-hydroxy-2-phenyl-ethyl)-amide,
- 3- Naphthalene-2-carboxylic acid [2-hydroxy-2-(4-nitro-phenyl)-ethyl]-amide,
- 4- Naphthalene-2-carboxylic acid [2-(4-chloro-phenyl)-2-hydroxy-ethyl]-amide,
- 5- Naphthalene-2-carboxylic acid [2-(4-bromo-phenyl)-2-hydroxy-ethyl]-amide,
- 6- 4-Chloro-N-(2-hydroxy-ethyl)-benzamide,
- 7- 4-Chloro-N-(2-hydroxy-2-phenyl-ethyl)-benzamide,
- 8- 4-Chloro-N-[2-hydroxy-2-(4-nitro-phenyl)-ethyl]-benzamide,
- 9- 4-Chloro-N-[2-(4-chloro-phenyl)-2-hydroxy-ethyl]-benzamide,
- 10- 4-Chloro-N-[2-(4-bromo-phenyl)-2-hydroxy-ethyl]-benzamide,
- 11- 4-Bromo-N-(2-hydroxy-ethyl)-benzamide,
- 12- 4-Bromo-N-(2-hydroxy-2-phenylethyl)-benzamide,
- 13- 4-Bromo-N-[2-hydroxy-2-(4-nitro-phenyl)-ethyl]-benzamide,
- 14- 4-Bromo-N-[2-(4-chloro-phenyl)-2-hydroxy-ethyl]-benzamide,
- 15- 4-Bromo-N-[2-(4-bromo-phenyl)-2-hydroxy-ethyl]-benzamide

The optimized geometries of the all molecules (1-15) were determined by using DFT/B3LYP method [20] with 6-31G(d,p) basis set. The optimized structures were then used to calculate some physicochemical properties, HOMO and LUMO energy, band gap energy ( $\Delta E$ ), electronegativity ( $\chi$ ), chemical hardness ( $\eta$ ), softness (S) and electronegativity index ( $\omega$ ) of all

molecules. All of these calculations were performed at the same level. All theoretical calculations in this work were carried out using Gaussian 09 program [21] and GaussView molecular visualization program [22]. In addition, the hyperpolarizability ( $\beta$ ) values of all molecules taken from literature [19]. All of these input and output values are listed in Table 2.

Table 2. HOMO and LUMO energies, the energy gap ( $\Delta E$ ), absolute electronegativity ( $\chi$ ) chemical hardness ( $\eta$ ), softness (S), electronegativity index ( $\omega$ ) and hyperpolarizability ( $\beta$ ) of the 15 different molecules using B3LYP/6-31G(d,p) level

Molecules	Inputs						Output	
	Reactivity Indexes							
	HOM O energies ( $E_{HOMO}$ )	LUM O energies ( $E_{LUMO}$ )	Ener gy gap ( $\Delta E$ )	Absolute electronegativity ( $\chi$ )	Chemical hardness ( $\eta$ )	Softness (S)	electronegativity index ( $\omega$ )	Hyperpolarizability ( $\beta$ )
1	- 5.984	- 1.414	4.57 0	-3.699	2.285	0.219	2.995	3.72

2	-	-	4.54		0.220	3.079	3.18
	6.011	1.469	2	-3.740	2.271		
3	-	-	3.75		0.266	4.920	12.38
	6.174	2.421	4	-4.298	1.877		
4	-	-	4.51		0.221	3.212	4.12
	6.066	1.550	5	-3.808	2.258		
5	-	-	4.54		0.220	3.170	4.15
	6.066	1.523	2	-3.794	2.271		
6	-	-	4.10		0.243	3.866	9.48
	6.038	1.931	7	-3.985	2.054		
7	-	-	4.16		0.240	3.868	8.27
	6.093	1.931	2	-4.012	2.081		
8	-	-	4.29		0.233	4.243	9.49
	6.419	2.122	8	-4.270	2.149		
9	-	-	5.16		0.193	3.136	5.27
	6.610	1.442	8	-4.026	2.584		
10	-	-	5.08		0.197	3.122	5.27
	6.528	1.442	6	-3.985	2.543		
11	-	-	5.38		0.186	2.969	5.48
	6.691	1.306	6	-3.998	2.693		
12	-	-	5.22		0.191	3.020	5.09
	6.582	1.360	2	-3.971	2.611		
13	-	-	4.46		0.224	4.964	12.10
	6.936	2.475	1	-4.706	2.230		
14	-	-	5.16		0.193	3.136	5.95
	6.610	1.442	8	-4.026	2.584		
15	-	-	5.05		0.198	3.160	5.95
	6.528	1.469	9	-3.998	2.530		

The GRA method was used to investigate which molecule would be more suitable for optoelectronic device applications. The method used is described below.

## 2.2. Gray Relational Analysis Method

Common methods such as linear programming and regression analysis are used in the solution of decision-making problems in the literature [23, 24]. For these methods, the number of sample data should be large. However, these methods seem to have difficulties in solving decision-making problems involving inefficient, unstable and irregular samples, and Gray Relational Analysis method, which takes gray theory into account, is used in such decision-making problems [25].

The Gray Theory was first proposed by Julong Deng in 1982 [26]. Gray theory, which provides modelling and solving problems that cannot be solved by stochastic or fuzzy decision-making methods; It is a frequently used method for inter-system analysis, model building, forecasting and decision-making problems. Looking at real life problems, it is very difficult to characterize all factors as fully positive or

fully negative. Likewise, it is not possible to evaluate all factors as completely specific or completely uncertain. If a system with no uncertainty is “white”, a system in which all factors are completely uncertain is “black”, partially definite and partially uncertain systems are defined as “gray” [27, 28]. The majority of events, processes, and processes in life are never all black or all white. We often build models and make decisions under incomplete, incomplete information and assumptions. The philosophy of the emergence of the gray system theory is also based on these ideas. System factors, the relationship between the factors, the structure of the system and the way it operates are the characteristic features of gray systems [29, 30]. When a problem consists of a single criterion, the alternative that is closest to the best value and farthest to the worst value that that criterion can have can be easily selected. However, when a problem consists of more than one criterion, it is not easy to detect it. This is where the GRA method comes into play. This method, which calculates the closeness of an alternative to the best value and the distance to the worst value in all criteria, succeeds in choosing the best alternative in total [28, 30]. GRA aims to solve

problems with inefficient sampling and poor information, and its basic principle is to use the similarity of geometric shape produced by different time series to reveal the relationship between them [31]. Today, this method is widely applied in many disciplines such as economics, sociology, industry, education, medical science, chemistry, geology, and aeronautics and space science [32].

The steps of the GRA method are detailed below [26];

*Step 1:* Preparing the data set and construct the decision matrix:

$$X = [x_1(1) \cdots x_1(n) \cdots x_m(1) \cdots x_m(n)] \quad (1)$$

*Step 2:* Construct the reference sequence and comparison matrix

$$x_0 = (x_0(j), \dots, x_0(n)) \quad j = 1, 2, \dots, n \quad (2)$$

*Step 3:* Normalize the data and construct the normalized matrix

If larger sequence values contribute positively, then normalization for “the larger the better” attributes is as follows:

$$x_i(k) = \frac{x_i^0(k) - \min x_i^0(k)}{\max x_i^0(k) - \min x_i^0(k)} \quad (3)$$

where  $x_i^0(k)$  is the original value in the row k. in the sequence i.  $x_i(k)$  is the value in the row k. in the sequence i after normalization,  $\min x_i^0(k)$  is the minimum value in the sequence i and  $\max x_i^0(k)$  is the maximum value in the sequence i.

If smaller sequence values contribute positively, then normalization for “the smaller the better” cost is as follows:

$$x_i(k) = \frac{\max x_i^0(k) - x_i^0(k)}{\max x_i^0(k) - \min x_i^0(k)} \quad (4)$$

Normalization for “the nominal the better” is as follows:

$$x_i(k) = 1 - \frac{|x_i^0(k) - x^0|}{\max x_i^0(k) - x^0} \quad (5)$$

where  $x^0$  is nominal value.

*Step 4:* construct the absolute value table and gray relational coefficient matrix.

Let k be the row k. on a n-length sequence and  $\varepsilon(x_0(k), x_i(k))$  be the grey relational coefficient at the point k. calculated using Equations (7), (8), (9) and (10).

$$\Delta = [\Delta_{01}(1) \cdots \Delta_{01}(n) \cdots \Delta_{0m}(1) \cdots \Delta_{0m}(n)] \quad (6)$$

$$\varepsilon(x_0(k), x_i(k)) = \frac{\Delta_{\min} + \xi \Delta_{\max}}{\Delta_{0i}(k) + \xi \Delta_{\max}} \quad (7)$$

$$\Delta_{0i}(k) = |x_0(k) - x_j(k)| \quad (8)$$

$$\Delta_{\min} = \min_j \min_k |x_0(k) - x_j(k)| \quad (9)$$

$$\Delta_{\max} = \max_j \max_k |x_0(k) - x_j(k)| \quad (10)$$

where  $\xi \in (0,1)$  is a coefficient between 0 and 1.

The function  $\xi$  sets the difference between  $\Delta_{0i}$  and  $\Delta_{\max}$ .

*Step 5:* The grey relational degree is calculated using Equation (11).

$$\gamma(x_0, x_i) = \frac{1}{n} \sum_{k=1}^n \varepsilon(x_0(k), x_i(k)) \quad (11)$$

If the weights of criteria are given in advance, grey correlation coefficients are calculated by the multiplying grey relationship coefficients and weights of the criteria.

$$\gamma(x_0, x_i) = \frac{1}{n} \sum_{k=1}^n \varepsilon(x_0(k), x_i(k)) \cdot (W_i(k)) \quad (12)$$

### 3. Findings

#### 3.1. Findings Obtained from Statistical Analysis

In this part of the study, the correlation analysis given in Table 3 was performed using the IBM SPSS Statistics 23 program to examine the effects of molecular reactivity indexes on hyperpolarization using the data obtained from the DFT study and given in Table 2.

Table 3. Correlation analysis

		$\beta$	$E_{\text{HOMO}}$	$E_{\text{LUMO}}$	$\Delta E$	$\chi$	$\eta$	S	$\omega$
$\beta$	Pearson	1	-.242	-	-.595*	-	-	.634*	.953**
	Sig. (2-tailed)		.385	.000	.019	.000	.019	.011	.000
$E_{\text{HOMO}}$	Pearson	-.242	1	.040	-.601*	.639*	-	.574*	-.116
	Sig. (2-tailed)	.385		.886	.018	.010	.018	.025	.682
$E_{\text{LUMO}}$	Pearson	-	.040	1	.775**	.795*	.775**	-	-
	Sig. (2-tailed)	.000	.886		.001	.000	.001	.000	.000
$\Delta E$	Pearson	-.595*	-	.775**	1	.231	1.000	-	-
	Sig. (2-tailed)	.019	.018	.001		.407	.000	.000	.002
$\chi$	Pearson	-	.639*	.795**	.231	1	.232	-.258	-
	Sig. (2-tailed)	.000	.010	.000	.407		.405	.353	.000
$\eta$	Pearson	-.595*	-	.775**	1.000*	.232	1	-	-
	Sig. (2-tailed)	.019	.018	.001	.000	.405		.000	.002
S	Pearson	.634*	.574*	-	-	-.258	-	1	.745**
	Sig. (2-tailed)	.011	.025	.000	.000	.353	.000		.001
$\omega$	Pearson	.953**	-.116	-	-	-	-	.745**	1
	Sig. (2-tailed)	.000	.682	.000	.002	.000	.002	.001	

\*\* . Correlation is significant at the 0.01 level (2-tailed).

\* . Correlation is significant at the 0.05 level (2-tailed).

When the result of the correlation analysis given in Table 3 is examined, with Hyperpolarizability;

- There is a significant and negative relationship at the 0.01 level between LUMO energies and absolute electronegativity,
- There is a significant and positive relationship at the 0.01 level between electronegativity index,
- There is a significant and negative relationship at 0.05 level between energy gap and chemical hardness,
- There is a significant and positive relationship at the 0.01 level between softness,
- There is no statistically significant relationship between HOMO energies,

was determined.

According to this result, the HOMO energies factor was not taken into account in determining which molecule would be more suitable for use in optoelectronic devices when the reactivity indexes were taken into account, using multi-criteria decision-making methods.

### 3.2. Findings from the GRA Method

In the first step of the method, the decision matrix and reference series in Table 4 were created using the data given in Table 2. While creating the reference series, LUMO energies, energy gap, absolute electronegativity and chemical hardness indexes were taken into account as benefit factors, while softness and electronegativity index were considered as cost factors, taking into account the result of correlation analysis.

Table 4. Decision matrix

	Reactivity Indexes					
	$E_{\text{LUMO}}$	$\Delta E$	$\chi$	$\eta$	S	$\omega$
Reference sequence	-2.475	3.754	-4.706	1.877	0.266	4.964
Molecule No	Min	Min	Min	Min	Max	Max
1	-1.414	4.57	-3.699	2.285	0.219	2.995
2	-1.469	4.542	-3.74	2.271	0.22	3.079
3	-2.421	3.754	-4.298	1.877	0.266	4.92
4	-1.55	4.515	-3.808	2.258	0.221	3.212
5	-1.523	4.542	-3.794	2.271	0.22	3.17
6	-1.931	4.107	-3.985	2.054	0.243	3.866
7	-1.931	4.162	-4.012	2.081	0.24	3.868
8	-2.122	4.298	-4.27	2.149	0.233	4.243

9	-1.442	5.168	-4.026	2.584	0.193	3.136
10	-1.442	5.086	-3.985	2.543	0.197	3.122
11	-1.306	5.386	-3.998	2.693	0.186	2.969
12	-1.36	5.222	-3.971	2.611	0.191	3.02
13	-2.475	4.461	-4.706	2.23	0.224	4.964
14	-1.442	5.168	-4.026	2.584	0.193	3.136
15	-1.469	5.059	-3.998	2.53	0.198	3.16

After the decision matrix was created, the data were normalized using equations (3) and (4). Here; softness and electronegativity index “bigger is better”, LUMO energies, energy gap, absolute

electronegativity and chemical hardness indexes were also evaluated as cost criteria and normalized as “lower is better”. The obtained normalized matrix is given in Table 5.

Table 5. Normalized matrix

	Reactivity Indexes					
	$E_{LUMO}$	$\Delta E$	$\chi$	$\eta$	S	$\omega$
Reference sequence	1.000	1.000	1.000	1.000	1.000	1.000
Molecule No	Min	Min	Min	Min	Max	Max
1	0.092	0.500	0.000	0.500	0.413	0.013
2	0.139	0.517	0.041	0.517	0.425	0.055
3	0.954	1.000	0.595	1.000	1.000	0.978
4	0.209	0.534	0.108	0.533	0.438	0.122
5	0.186	0.517	0.094	0.517	0.425	0.101
6	0.535	0.784	0.284	0.783	0.713	0.450
7	0.535	0.750	0.311	0.750	0.675	0.451
8	0.698	0.667	0.567	0.667	0.588	0.639
9	0.116	0.134	0.325	0.134	0.088	0.084
10	0.116	0.184	0.284	0.184	0.138	0.077
11	0.000	0.000	0.297	0.000	0.000	0.000
12	0.046	0.100	0.270	0.100	0.063	0.026
13	1.000	0.567	1.000	0.567	0.475	1.000
14	0.116	0.134	0.325	0.134	0.088	0.084
15	0.139	0.200	0.297	0.200	0.150	0.096

In the next step, the absolute values were calculated with the equation (8) and the gray relational

coefficient matrix given in Table 6 was obtained by using the equations (7), (9), (10) and (11).

Table 6. Gray relational coefficient matrix

Molecule No	Reactivity Indexes					
	$E_{LUMO}$	$\Delta E$	$\chi$	$\eta$	S	$\omega$
1	0.355	0.500	0.333	0.500	0.460	0.336
2	0.367	0.509	0.343	0.509	0.465	0.346
3	0.915	1.000	0.552	1.000	1.000	0.958
4	0.387	0.517	0.359	0.517	0.471	0.363
5	0.380	0.509	0.356	0.509	0.465	0.357
6	0.518	0.698	0.411	0.697	0.635	0.476
7	0.518	0.667	0.420	0.667	0.606	0.476
8	0.623	0.600	0.536	0.600	0.548	0.580
9	0.361	0.366	0.425	0.366	0.354	0.353
10	0.361	0.380	0.411	0.380	0.367	0.351
11	0.333	0.333	0.416	0.333	0.333	0.333

12	0.344	0.357	0.407	0.357	0.348	0.339
13	1.000	0.536	1.000	0.536	0.488	1.000
14	0.361	0.366	0.425	0.366	0.354	0.353
15	0.367	0.385	0.416	0.385	0.370	0.356

The Pearson Correlation coefficients obtained from the correlation analysis given in Table 3 were used to determine the weights of the reactivity indexes. Here,

the absolute values of the Pearson Correlation coefficients were normalized and the weights given in Table 7 were obtained.

Table 7. Weights of the reactivity indexes

	$E_{LUMO}$	$\Delta E$	$\chi$	$\eta$	S	$\omega$
Pearson Correlation coefficient	0.935	0.595	0.867	0.595	0.634	0.953
Normalized values (weights)	0.204	0.130	0.189	0.130	0.139	0.208

Then, using equation (12), the weighted gray relational degrees given in Table 8 were obtained.

Then, the molecules were ranked according to the total gray relational degrees.

Table 8. Weighted gray relational degree and ranking

Molecule No	Reactivity Indexes						Total gray relational degree	Ranking
	$E_{LUMO}$	$\Delta E$	$\chi$	$\eta$	S	$\omega$		
1	0.073	0.06	0.063	0.065	0.064	0.070	0.0570	9
2	0.075	0.06	0.065	0.066	0.064	0.072	0.0584	8
3	0.187	0.13	0.105	0.130	0.138	0.199	0.1270	1
4	0.079	0.06	0.068	0.067	0.065	0.076	0.0603	6
5	0.078	0.06	0.067	0.066	0.064	0.074	0.0594	7
6	0.106	0.09	0.078	0.091	0.088	0.099	0.0788	4
7	0.106	0.08	0.080	0.087	0.084	0.099	0.0774	5
8	0.127	0.07	0.101	0.078	0.076	0.121	0.0831	3
9	0.074	0.04	0.081	0.048	0.049	0.073	0.0531	12
10	0.074	0.04	0.078	0.049	0.051	0.073	0.0535	11
11	0.068	0.04	0.079	0.043	0.046	0.069	0.0498	14
12	0.070	0.04	0.077	0.046	0.048	0.071	0.0513	13
13	0.204	0.07	0.189	0.070	0.068	0.208	0.1155	2
14	0.074	0.04	0.081	0.048	0.049	0.073	0.0531	12
15	0.075	0.05	0.079	0.050	0.051	0.074	0.0542	10

In Table 8, according to the total gray relational degrees, the 3rd molecule (*Naphthalene-2-carboxylic acid [2-hydroxy-2-(4-nitro-phenyl)-ethyl]-amide*) ranked 1st with a gray relational degree of 0.1270, and the 13th molecule (*4-Chloro-N-[2-hydroxy-2-(4-nitro-phenyl)-ethyl]-benzamide*) 2nd and 11th molecules (*4-Chloro-N-(2-hydroxy-ethyl)-*

*benzamide*) ranked in last with a gray relational grade of 0.0498.

Finally, the results obtained based on the hyperpolarizability values obtained by DFT analysis and the total gray relational degree obtained from the GRA were compared and given in Table 9.

Table 9. Comparison of results from DFT and GRA

Molecule	DFT Analysis		GRA	
	Hyperpolarizability	Ranking	Total gray relational degree	Ranking
1	3,72	12	0,0570	9
2	3,18	13	0,0584	8
3	12,38	1	0,1270	1
4	4,12	11	0,0603	6
5	4,15	10	0,0594	7
6	9,48	4	0,0788	4
7	8,27	5	0,0774	5
8	9,49	3	0,0831	3
9	5,27	8	0,0531	12
10	5,27	8	0,0535	11
11	5,48	7	0,0498	14
12	5,09	9	0,0513	13
13	12,10	2	0,1155	2
14	5,95	6	0,0531	12
15	5,95	6	0,0542	10

When Table 9 is examined, it is seen that the molecules in the top 5 in both DFT and GRA are the same. This result shows that the results from DFT and GRA support each other. Therefore, it can be said that the 3rd molecule (*Naphthalene-2-carboxylic acid [2-hydroxy-2-(4-nitro-phenyl)-ethyl]-amide*), which ranks first in both DFT and GRA, is a suitable molecule that can be used in optoelectronic device applications.

#### 4. Discussion and Conclusion

In this study, theoretical calculation of the frontier molecular orbitals (HOMO and LUMO), band gap energy ( $\Delta E$ ), and reactivity indexes, electronegativity ( $\chi$ ), chemical hardness ( $\eta$ ), softness (S) and electronegativity index ( $\omega$ ) of some molecules. Molecular properties have been calculated by using the DFT method with 6-31G(d,p) basis set. The molecule 3 shows the most nonlinear optical property among all molecules. Statistical analysis was performed between the reactivity indices and hyperpolarization values obtained by DFT, and the appropriate molecule that could be used in electro optic devices was determined. According to the

results obtained from the statistical analysis of the study, it was determined that the electronegativity index affected hyperpolarizability the most, and there was no significant relationship between HOMO energy and hyperpolarizability. In the second stage of the study, according to the results obtained from GRA, it was determined that the 3rd molecule (*Naphthalene-2-carboxylic acid [2-hydroxy-2-(4-nitro-phenyl)-ethyl]-amide*) was the most suitable molecule to be used in electro optic device applications. The highest hyperpolarization value in this molecule, which gives the best results, is thought to be due to the fact that it has pi electrons of naphthalene and nitrophenyl groups that can be conjugated. When the DFT and GRA results were compared, it was determined that the top 5 most suitable molecules were the same in both analyses. Considering this comparison result, it was concluded that multi-criteria decision-making methods can be used in these analyses. In future studies, the hyperpolarizability value can be estimated by using the reactivity index values in machine learning algorithms.

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