

Heuristic Approach to Comparing the Environmental Impacts of Carbon Nanotube Production Methods

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ABSTRACT

Carbon Nanotubes (CNTs) production so far has its own advantages and disadvantages. Some methods that can be used in producing CNTs are chemical vapor deposition (CVD), laser ablation, and arc discharge. The three methods have their own requirements, this causes different environmental impacts on each method. Studies into the environmental impact of the CNTs production process found that during thermal pretreatment of the reactant gas, more than 45 by-products were formed, including methane, volatile organic compounds, and polycyclic aromatic hydrocarbons. Calculating the environmental impact of CNTs production method often has challenges in implementation, because each production process has different systems and needs. One way to overcome this problem is by using the heuristic method for forecasting environmental impact, which can be done with the Multi-Criteria Decision Analysis algorithm. The method can calculate uncertainty in each scenario, by normalizing the given load value. In this study, the CVD method has the best solution and objective value results compared to laser ablation and arc discharge. The best solution and objective values that show the value of scenario quality and environmental impact in each method, in CVD the solution obtained in the 34th generation has an epsilon value of 0.00251. The generation shows the performance of the scenario, while the epsilon value shows the value of the environmental impact, the smaller the generation, the better the scenario, while the smaller the epsilon value, the smaller the environmental impact.

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Keywords: Arc discharge, carbon nanotubes, CVD, environmental impact, heuristic, laser ablation, multi-criteria decision analysis

I. Introduction

Production of carbon nanotubes (CNTs) has seen significant advancements in recent years due to their unique properties and wide applications in various fields, including electronics, composites, and medicine. However, the environmental impacts of CNTs production remain a concern due to the energy consumption, raw material use, and waste generation associated with these processes [1]. The production methods for CNTs such as chemical vapor deposition (CVD), laser ablation, and arc discharge are well-documented in the scientific literature [2]. These methods each have their own advantages and disadvantages, which can significantly influence the environmental impacts of CNTs production [3].



One study evaluated the potential environmental impacts of CNTs synthesis by CVD. The study found that during the thermal pretreatment of the reactant gases, over 45 side products were formed, including methane, volatile organic compounds (VOCs), and polycyclic aromatic hydrocarbons (PAHs) [4]. This finding suggests several environmental concerns with the existing process, including potential discharges of the potent greenhouse gas, methane (up to 1.7%), and toxic compounds such as benzene and 1,3-butadiene (up to 36000 ppmv) [5]. From the impact analysis of CNTs production methods with CVD conducted by previous researchers, it is necessary to compare the environmental impact caused by other methods. Despite the growing body of research on CNTs production and its environmental impacts, there is still a lack of comprehensive comparative studies on the environmental impacts of different CNTs production methods [6]. This study aims to fill this gap by employing a heuristic approach to compare the environmental impacts of various CNTs production methods. This research is important for the development of sustainable and green production methods of CNTs, as shown in Figure 1, so as to reduce the environmental impact caused by the production of CNTs.

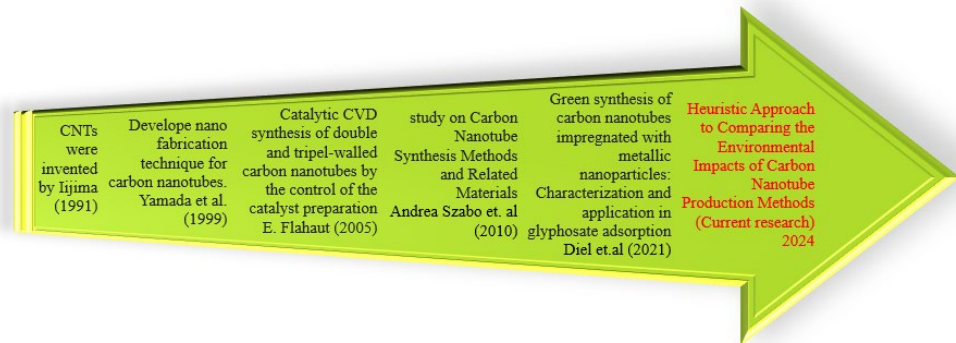


Fig. 1. State of the art study

Production of CNTs involves various methods, including CVD, metal organic CVD (MOCVD), plasma enhanced CVD (PECVD), laser ablation, and arc discharge [4]. CVD uses a carbon precursor that reduces in a reaction chamber to form single-walled CNTs (SWCNTs) [5]. MOCVD employs a metal organic compound to reduce the carbon precursor alongside it, leading to multi-walled CNTs (MWCNTs). PECVD similarly introduces a carbon precursor that is reduced by plasma to create MWCNTs. Laser ablation offers precision control over the growth environment and enables the formation of high-quality CNTs arrays with controlled orientation and diameter. Arc discharge, while less precise, creates plasma between electrodes to serve as a reducing agent, which is suitable for large-scale production due to its speed and lower cost [6]. Each method has its advantages and is chosen based on the desired properties of the resulting CNTs and the scale of production required.

CVD, laser ablation, and arc discharge are key techniques for synthesizing nanomaterials such as carbon nanotubes (CNTs). CVD is a bottom-up process that enables the creation of dense films or particles with high purity, utilizing a carbon precursor that reduces within a reaction chamber. This method is particularly effective for producing single-walled CNTs (SWCNTs) and depositing various thin films. While CVD is robust, it requires careful control over the process to achieve the desired properties [7]. Laser ablation, known for its precision, uses a laser to generate plasma in a gas mixture, forming high-quality CNTs arrays with controlled dimensions and orientation, which is crucial for

applications demanding uniform structures. Arc discharge, despite being less precise, generates plasma between electrodes for reduction, making it suitable for mass production due to its efficiency and cost-effectiveness, though it may compromise the quality of the CNTs [8]. Each of these methods has its advantages and disadvantages, and the choice of method can significantly influence the environmental impacts of CNTs production [8].

Table 1. Advantages and disadvantages CNTs production method.

Production Method	Advantages	Disadvantages
CVD	High yield, good control on diameter and length, easy to scale up [9].	High temperatures, expensive equipment, careful control of operation [10].
MOCVD	Good control over diameter and length, high yield, less expensive than CVD [5].	Requires careful control of operating conditions and expensive equipment, less scalable than CVD [11].
Laser ablation	Low metallic impurities. higher yield of SWCNTs, better properties and narrower sizes compared to laser ablation method. More efficient with certain catalysts such as Ni, Co, Fe, and Pt [12].	Not economically advantageous because the procedure requires high-purity graphite rods and significant laser power. The obtained CNTs are not uniformly straight but instead contain some branching [13].
PECVD	High yield, good control over diameter and length, less expensive than CVD and MOCVD [14].	Requires careful control of operating conditions, requires expensive equipment, less scalable than CVD and MOCVD [15].
Diffusion flame synthesis	Low cost, simple procedure, good control over diameter and length [16].	need high temperatures, low yield, difficult to control diameter and length [17].
Electrolysis	Less expensive than CVD, good control over diameter and length [18].	Requires high voltages, low yield, difficult to control diameter and length [19].
Solar energy	Renewable energy source, low cost, good control over diameter and length [20].	Requires special equipment, low yield, difficult to control diameter and length [21]
Arc discharge	Produces bulk CNTs, fewer structural defects. Most of the synthesized CNTs in arc discharge are perfectly straight [22].	Little control effect over the CNTs alignment. Purification of the obtained CNTs is necessary [23].
Heat treatment of a polymer	Low cost, simple procedure, good control over diameter and length [5].	Requires high temperatures, low yield, difficult to control diameter and length [5].
Low-temperature solid pyrolysis	Low cost, simple procedure, good control over diameter and length [24].	need special equipment, low yield, difficult to control diameter and length [25].

The method used in the CNTs production process is influenced by several parameters namely temperature, energy consumption, design, raw material use and waste generation, cnt yield, potential health risks. These parameters will be analyzed regarding the effect on environmental pollution caused by life cycle assessment (LCA). LCA is a methodology that

analyzes the environmental aspects and potential impacts associated with a product, process, or service system from cradle to grave [26]. It is widely used in the field of sustainability to evaluate the environmental performance of different alternatives and make informed decisions. Multiple criteria decision analysis (MCDA) is another approach used in decision-making processes [27]. MCDA has been applied in various fields, such as environmental management, urban planning, and industrial engineering [28]. However, it's worth noting that the application of MCDA in LCA requires careful consideration of the choice of criteria, weight assignment, and normalization method [29]. These factors can significantly influence the results of the analysis and, therefore, need to be handled with care.

Heuristic models in LCA are computational tools used to simplify the complexity of LCA and make it more manageable. They are particularly useful in situations where it is not feasible or practical to perform a complete LCA due to the complexity of the system under study or the availability of incomplete or uncertain data [30]. In the context of CNTs production, heuristic models can be used to estimate the environmental impacts of different production methods. For instance, a heuristic model could be used to estimate the energy consumption, raw material use, and waste generation associated with each method based on available data and assumptions [31]. This can provide a rough estimate of the environmental impacts without requiring a detailed inventory analysis or impact assessment [32]. However, there are several limitations to using heuristic models in LCA. First, they rely heavily on assumptions and may not accurately represent the true environmental impacts if these assumptions are not valid. Second, they may not capture all the relevant environmental impacts, especially those that are indirect or less obvious [33]. Third, they may not be suitable for all types of systems or all types of impacts. Despite these limitations, heuristic models can still be valuable tools in LCA, particularly in the context of CNTs production [34]. They can provide quick and easy estimates of environmental impacts, which can be useful for decision-making and policy-making purposes [35]. They can also serve as a starting point for more detailed LCA studies, providing insights into the key factors affecting the environmental impacts of a system [36].

II. Methods

Defining criteria is the first step considered in the MCDA scenario [37]. These criteria include factors that affect CNTs production such as energy consumption, CNTs yield, process properties, reaction temperature, nanotube selectivity, process parameter control, carbon source, and purification [39]. Each of these factors will represent criteria in the MCDA problem. Extensive data collection, analysis, and review were conducted against existing literature studies on CNTs production methods to ascertain the weight of influential factors. The stages of the method used are shown in Figure 2.

A. Problem Identification and Literature Study

Main problems of CNTs production that can cause environmental damage are analyzed to be used as parameters in comparing and forecasting the environmental impact of CNTs production. Data obtained from references and previous research on CNTs production, especially in CVD, laser ablation, and arc discharge methods, were weighted to give a fair value to each method [37]. Furthermore, the heuristic method was used to forecast the environmental impact of CNTs production methods.

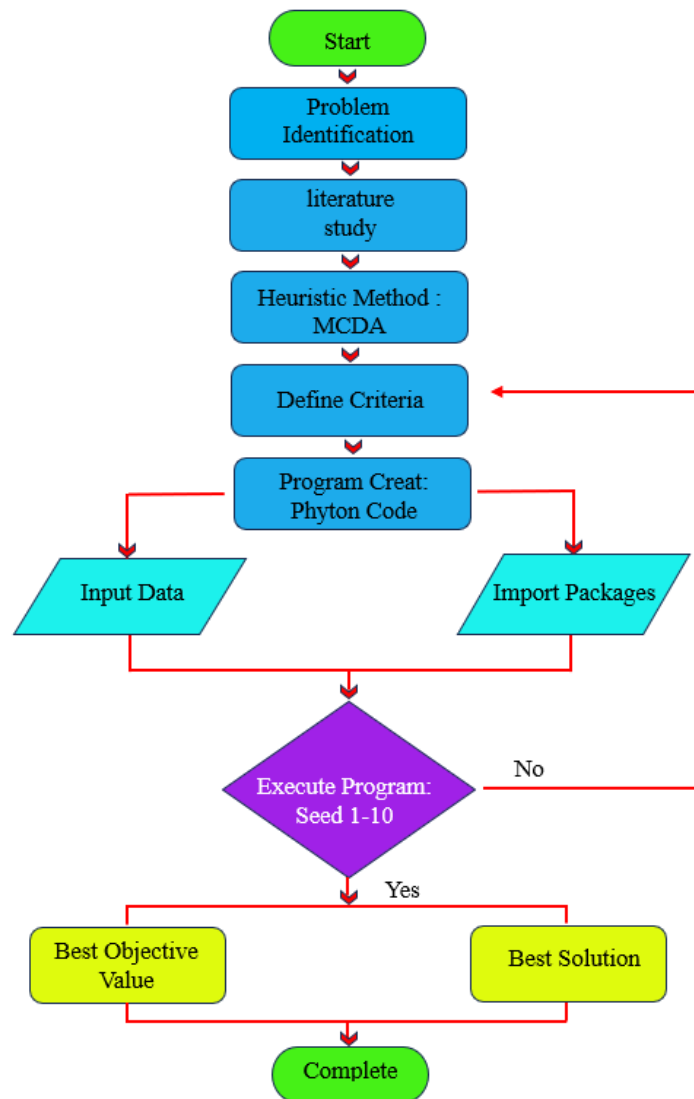


Fig. 2. Flowchart forecasting environmental impact with Heuristic MCDA method

B. Heuristic Method and Define Criteria

The heuristic method uses expert knowledge and historical data to identify key criteria and sub-criteria relevant for the environmental impact of CNTs production. These criteria include greenhouse gas emissions, energy consumption, pollution, and solid waste generated from CNTs production metrics [38]. The heuristic method used was MCDA method.

C. Program Creat

1. Input Data and Import Packages

Data that has met the criteria will be input into the program to be processed in the MCDA method. The Numpy and Pymoo packages are used as Python code libraries that function for multi-objective modeling and optimization in the MCDA algorithm[40]. Numpy is used for processing numerical arrays and manipulating numerical data related to MCDA problems, such as decision matrices and goal value vectors Pymoo is used to represent multi-objective optimization problems, in the context of MCDA which includes criteria or objectives to be predicted and optimized [41].

2. Define Weights

Assigning weights to each criterion according to the impact of the factor on the environment [42]. Decision making on each factor's loading with another factor needs to be analyzed with one another. This weight will determine the relative impact of each criterion in the decision-making process to determine the weight of each factor[43]. Loading needs to be defined specifically because it is carried out quantitatively in the MCDA problem.

3. Define Problem

The MCDA method has a class that inherits from the "problem" class. The problem class is used to define optimization problems [44]. The package problem used in the MCDA method is pymoo. The problem with the MCDA method needs to be initialized to determine its characteristics. The characteristics of MCDA include the number of decision variables in the MCDA scenario which is 10. Scenarios are controlled by seeds where there are 10 seed variations symbolized by numbers 1-10, the number of objectives which is 2, namely best objective value and best solution. The best objective value represents the environmental impact value of the 3 CNTs production methods and the best solution represents the best scenario used to predict environmental impact. the lower and upper bounds of the value of each decision variable which is 0 to 1, and the evaluation applied elementally to the input is "True".

4. Calculate MCDA Problem

The quantitative operation of the MCDA problem is modeled in the Weighted Sum Model (WSM). This model is used to combine a number of different criteria or attributes into a single value, which is referred to as weighted value or utility value[45]. In the context of decision optimization or evaluation, the main objective of this model is to find a combination of decision variables that provide maximum or minimum weighted value, depending on the type of problem at hand. The objective function is defined in the evaluation and represents the load amount model used in the MCDA method. The goal value is calculated as the sum of the multiplication of elements of the decision variable and a load [46]. The value of the weight or utility to be optimized ($F(x)$) can be found by multiplying the number of criteria (n) by the value of the decision variable (x_i) and the weight given to each criterion (w_i).

$$F(x) = \sum_{i=1}^n w_i \cdot x_i \dots\dots\dots (1)$$

The context of the MCDA problem is to find a combination of decision variable values (x) that optimizes weighted values ($F(x)$), where weights (w_i) are parameters that can be adjusted according to the preferences or interests of decision makers.

5. Define and perform Optimization NSGA II

The NSGA-II (Non-dominated Sorting Genetic Algorithm II) is a popular multi-objective optimization algorithm often used in (LCA). This algorithm is known for its high computational efficiency, ability to create good solutions, and maintenance of population diversity through a mechanism known as crowding comparison. In the context of LCA, NSGA-II has been used to optimize the design of buildings to minimize life cycle costs and CO₂ emissions. A study used NSGA-II in conjunction with an artificial neural network (ANN) model to identify optimal design solutions. The ANN model was trained and validated using simulation data and then used as the evaluation function for the multi-objective optimization algorithm. The results demonstrated potential reductions in life cycle

cost and CO₂ emissions compared to the initial design, indicating that the optimization approach improved building performance [35]. This application of NSGA-II in LCA shows how it can be used to find trade-off solutions that satisfy multiple conflicting objectives, such as cost and environmental impact. The use of ANNs in conjunction with NSGA-II further enhances the capabilities of the algorithm, allowing it to handle complex, non-linear problems and speed up the convergence process [35]. NSGA-II allows researchers to model and evaluate various CNTs production scenarios under different conditions. This can include scenarios with different raw materials, new production technologies, or different environmental policies.

NSGA II is used to find optimal solutions to multi-objective problems [47]. The MCDA problem uses NSGA II as an initialization algorithm, namely to determine the population of individuals in each generation that is 1000; Crossover probability determines how often crossovers will be assigned to pairs of reproductive individuals that are set at 0.9. Mutation probability determines how often mutations will be applied to each decision variable in an individual, which is 1 according to the maximum value of loading. The number of generations determined based on termination is set according to the number of generations in the algorithm that has reached the convergence of diversity, the generation algorithm will stop after reaching the 1000th generation.

The performance of the algorithm starts from the function to start the optimization process, optimization problems that have been previously defined using MCDA Problems, optimization algorithms, and termination criteria that will determine when optimization is stopped [48]. Seed is used to reproduce results by assigning values to random number generators. Seeds can be changed at each execution to find the best generation potential, the best illusion and the best objective value from the scenario created [49]. The optimization process is displayed using "verbose=True" to display information during execution.

The determination of the number of generations, crossovers, and mutations is based on the Grid Search and Bayesian Optimization experimental approaches. The Grid Search approach is used to test different parameter combinations and evaluate the performance of each combination based on relevant metrics such as convergence and solution diversity. Bayesian Optimization is used to select parameters based on the performance of previous iterations. For example, if the previous iteration has converged at the 100th generation, then that generation can be set for the next MCDA scenario.

NSGA II was chosen because there are two conflicting objectives, namely between the best objective value and the best solution. In each scenario run, the value of the two objectives is not always directly proportional. Because both objectives have different influence variables. Best solution is influenced by variables from the algorithm scenario and the best objective value of the CNTs production metric.

D. Best Solution and Best Objective

The results produced by the scenario designed in this study with the MCDA method are divided into two, namely Best Solution and Best Objective value. "Best Solution" takes the best solution from the results of MCDA optimization. This solution is a combination of values from several alternatives that meet the best criteria according to the scenario created in the MCDA method [50].

"Best Objective Value" takes the best objective value associated with the best solution in the MCDA method. The objective value reflects how well the solution meets the criteria

or preferences set [41]. The preference set in the MCDA scenario of this study is that the smaller the value of "Best Objective Value", the better the method used.

III. Results and Discussions

Determining environmental impact considers the inputs and outputs of a resulting system. Environmental impact forecasting using MCDA Heuristics NSGA II algorithm requires factors that affect the process of making carbon nanotubes, in the forecasting process involves several input, output, and process control factors. The factors considered can be seen in Table 2.

Table 2. Data comparison CVD, laser ablation, and arc discharge [8].

Process	CVD	Laser ablation	Arc discharge
Temperature	500-1100°C	3000°C	3000-4000°C
Energy	Low	High	High
Nanotube selectivity	High	Low	Low
Source of carbon	Easy available	Difficult	Difficult
Purification of CNT	Low	High	High
CNTs yield	95-99%	70%	30%
Process nature	Continuous	Batch	Batch
Process parameter control	Low	High	High
Nanotube graphitization	Middle	High	High

Table 2 data can generate the load value of each variable by considering from other method variables. The value of the load is assumed by level, for example in the energy variable it is assumed with two levels, namely level 1 which defines "Low" and level 2 defines "High" in Table 2. Converting data variables to load values is shown in Table 3.

There are special cases of variable temperature, nanotube graphitization, and CNTs yield. The variable CNTs yield uses percent in the assessment, thus indicating the much CNTs production produced on each method. The determination in Table 3 of CNTs variable variables shows the residual value of products that cannot become carbon nanotubes. In variable temperature and nanotube graphitization there are three levels, namely low, medium, and high, so that the determination of the loading value is adjusted between numbers 1-3.

Table 3. Weight data comparison CVD, laser ablation, and arc discharge.

Process	CVD	Laser ablation	Arc discharge
Temperature	1	2	3
Energy	1	2	2
Nanotube selectivity	2	1	1
Source of carbon	1	2	2
Purification of CNT	2	1	1
CNTs yield	0.03	0.3	0.7
Process nature	1	2	2
Process parameter control	2	1	1
Nanotube graphitization	2	3	3

The value obtained by adjusting the level according to the variable in each factor will be processed again using the normalized method. The goal is to normalize the value of the loading factor in CVD, laser ablation, and arc discharge methods. The data that has been processed is the loading value that will be entered into the environmental forecasting impact using the MCDA heuristic method. The loading on CNTs yield is based on the percentage of the three different methods on the CNTs production parameters indicated by nominal numbers. The normalized loading values are obtained in Table 4.

Table 4. Normalize data comparison CVD, laser ablation, and arc discharge.

Process	CVD	Laser ablation	Arc discharge
Temperature	0.17	0.33	0.50
Energy	0.20	0.40	0.40
Nanotube selectivity	0.50	0.25	0.25
Source of carbon	0.20	0.40	0.40
Purification of CNT	0.50	0.25	0.25
CNTs yield	0.03	0.29	0.68
Process nature	0.20	0.40	0.40
Process parameter control	0.50	0.25	0.25
Nanotube graphitization	0.25	0.375	0.375

The normalized data is a factor that affects the results of the predicted environmental impact value, this needs to be done so that the assessment is more objective at the specified scope. Normalizing data in the context of MCDA and LCA involves standardizing the values of different variables so they fall within a common scale. This process is crucial because it allows for comparison across different parameters and contributes to the overall accuracy and reliability of the assessment. In the field of LCA, normalization is often used during the development of weighting factors. As per the work of Prado, these weighting factors play a significant role in determining the importance of different impact categories in the normalize[48]. Therefore, normalization helps in ensuring that these weights reflect the relative importance of various impacts accurately.

However, the normalization process is not without its challenges. According to Baran-kooiker, the normalization process can significantly affect the weighting results in LCA. They found that the choice of normalization method and the inclusion of background information could influence the weighting [49]. Another critical aspect of normalization is its interaction with other MCDA methods. For instance, Domingues explored the future directions of normalization and weighting in LCA, indicating that these processes may need to evolve alongside advancements in other areas of MCDA [50]. Despite these complexities, normalization remains a key step in the MCDA and LCA process. It provides a consistent scale for comparing different variables, thereby enabling more accurate and reliable assessments. Future research should continue to explore and refine this process, taking into account the latest developments in MCDA and LCA.

The convergence of the tested data using normalized load values can be seen in Figure 3. The graph shows that the forecasting results of the three methods began to converge in generation <100, determining that repeating 150 calculations has produced convergent data. When repeating calculations in all three methods converges. Ideal and false values for each generation performed can be known, The goal is to see how much ideal data is used to determine the value of environmental impact.

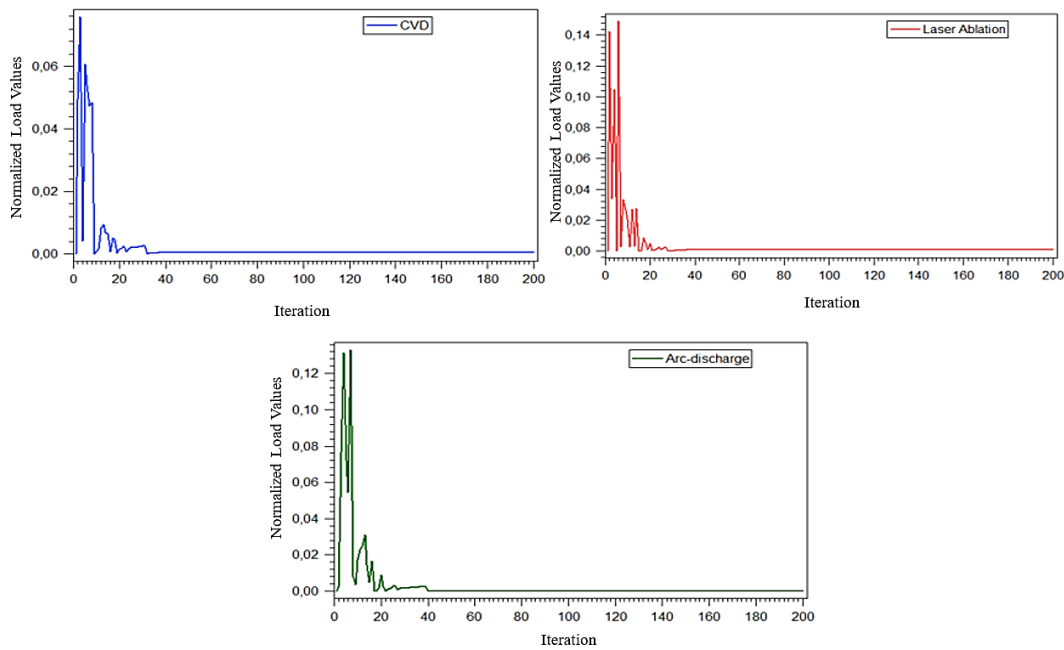


Fig. 3. Convergence forecasting heuristic MCDA

Environmental impact factors that have been run with the NSGA II algorithm in the MCDA method produce epsilon values that indicate ideal or false values in generation. The ideal indicator is shown in Figure 4, namely in generation 39 with an epsilon value of 0.00255 in the arc discharge method, generation 34 with an epsilon of 0.00251 in the CVD method, generation 34 with an epsilon 0.00262 in the laser ablation method. The performance indicator stays at ideal for many generations. This suggests that the algorithm is performing well and continuously finding better solutions. However, toward the end of the optimization process, the performance indicator switches to f, indicating that the optimization process is stagnating. The stagnation is caused by a lot of generation so that crossover probability and mutation probability have reached a concurrent process and caused the best solution found 0. Performance indicators of the MCDA method can be known from the diversity value run by the algorithm. The sooner the data finds which last ideal value, the better performance will be [51].

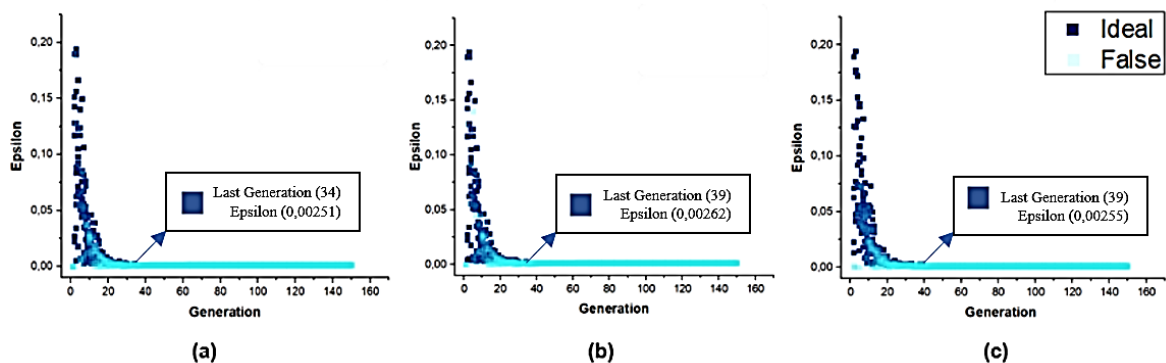


Fig. 4. Diversity forecasting heuristic MCDA of (a) CVD, (b) Laser ablation, and (c) Arc discharge

Effect of performance is influenced by the load value and scenario results of the three methods. A minimum loading value will provide a faster and better performance, as in Figure 3, which shows that the CVD method has the best performance. Once normalized,

CVD has the smallest dominant loading value among other methods. The uncertainty of the combination of load values has been identified in Figure 3, where ideal and false values are known. It can be concluded that because the scenario created is minimized, the result of the minimum loading value will have the fastest performance and the lowest scenario value.

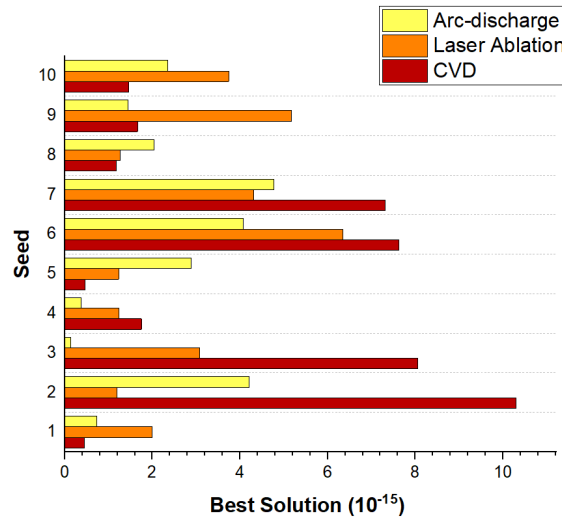


Fig. 5. Best solution forecasting heuristic MCDA

Seeds are used to reproduce results randomly so that seeds can be validated by changing the value in each forecasting calculation [52]. The best solution is the value of the scenario used in the three methods; the result value of the best solution can be seen in Figure 5. The graph shows that there are two methods that have the best value of 40% each, namely CVD and arc discharge. The percentage value is determined based on the number of best scenarios obtained from the forecasting process 10 times from three methods. CVD method scenarios have the best value of 4, namely at seed 1 (0.45×10^{-15}), 5 (0.46×10^{-15}), 8 (1.17×10^{-15}), and 10 (1.46×10^{-15}). While the arc discharge method has the best solution value of 4 at the 3rd seed (0.14×10^{-15}), 4th (0.37×10^{-15}), 6th (4.08×10^{-15}), and 9th (1.45×10^{-15}). Laser ablation has the best solution value of at least 20% at seed 2 (1.19×10^{-15}) and 7 (4.31×10^{-15}). This shows that the CVD and arc discharge methods have the best variable scenario values among the three methods.

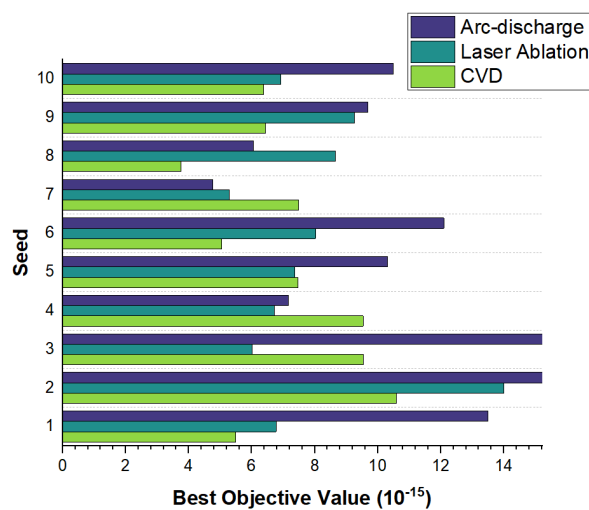


Fig. 6. Best objective value forecasting heuristic MCDA

Figure 6 shows the graph of the best objective value of the three methods of producing carbon nanotubes; the best objective value is the value that represents the environmental impact of the variability that has been normalized before. Variations in scenarios, loadings, and seeds performed will result in different environmental impacts in each calculation. The graph shows that the CVD method has the best value of 60% compared to the laser ablation and arc discharge methods. The percentage value is obtained from the sum of the best objective values obtained from the MCDA forecasting process, namely 6 of the 10 scenarios run on the three methods. The best objective values of the CVD method are located at seeds 1 (5.48×10^{-15}), 2 (10.6×10^{-15}), 6 (5.05×10^{-15}), 8 (3.76×10^{-15}), 9 (6.44×10^{-15}), and 10 (6.38×10^{-15}). This shows that despite the seed changes that can affect random load generation, the CVD method still has the best environmental impact value compared to laser ablation and arc discharge; seed changes made still produce superior values from the CVD method because the program has converged [53].

IV. Conclusions

Production of CNTs will have an environmental impact, so it is necessary to calculate the environmental impact on the various methods used to produce CNTs. Some of the methods used in producing CNTs are CVD, laser ablation, and arc discharge. The environmental impact can be predicted by using the MCDA heuristic method. The value can be known by giving the normalized weight value to each influencing factor into the MCDA method.

The results of forecasting the environmental impact value show that the CVD method has the lowest value. Evidenced by the ideal last generation of the CVD method is in the 34th generation and has an epsilon value of 0.00251, this result is the fastest generation and the lowest epsilon value among other methods. In addition, 40% of the best solution data and 60% of the best objective value data show that the CVD method has a low environmental impact value compared to the laser ablation and arc discharge methods. By comparing the environmental impact values of the three methods, CVD is the most environmentally friendly method in producing CNTs. The results of this comparison of CNTs production methods in the future are expected to help the industrial world in the selection of CNTs production methods to produce CNTs that are sustainable and environmentally friendly and contribute to reducing environmental damage caused by the potential discharge of strong greenhouse gases, methane, and toxic compounds caused by the CNTs production process. This study on the environmental impact of CNTs production still has shortcomings, because the forecasting carried out is based on the loading value and depends on previous references and research, so experiments are needed to find out more specifically the environmental impact of various CNTs production methods.

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