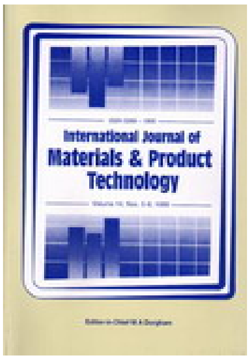

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Muath Jarrah, Zakaria N.M. Alqattan, Abdul Syukor Mohamad Jaya, Sharif Naser Makhadmeh, Ahmed Ismail Abu-Khadrah, Ibrahim Aljarrah, Osama Ahmad Alomari

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Improving CNTs properties using computational intelligence algorithms

Muath Jarrah*

College of Computing and Informatics,
University of Sharjah,
Sharjah, 27272, UAE
and
Department of Computer Science,
University Malaysia of Computer Science and Engineering (UNIMY),
Cyberjaya, Selangor, Malaysia
Email: aljarrahmuath@gmail.com
*Corresponding author

Zakaria N.M. Alqattan

Department of Cyber Security and Cloud Computing Techniques Engineering,
Northern Technical University,
Mosul, Iraq
and
Department of Computer Science,
Kulliyyah of Information and Communication Technology,
International Islamic University Malaysia (IIUM),
Kuala Lumpur, Malaysia
Email: Zakaria@ntu.edu.iq

Abdul Syukor Mohamad Jaya

Department of Intelligent Computing and Analytics,
Universiti Teknikal Malaysia Melaka,
Hang Tuah Jaya, 76100, Melaka, Malaysia
Email: syukor@utem.edu.my

Sharif Naser Makhadmeh

Department of Data Science and Artificial Intelligence,
University of Petra, Amman,
11196, Jordan
Email: sharif.makhadmeh@uop.edu.jo

Ahmed Ismail Abu-Khadrah

College of Computing and Informatics,
Saudi Electronic University,
Riyadh, Saudi Arabia
Email: a.abukhadrah@seu.edu.sa

Ibrahim Aljarrah

Department of Computer Information Systems (CIS),
Jordan University of Science and Technology,
Irbid, Jordan
Email: imaljarrah18@cit.just.edu.jo

Osama Ahmad Alomari

Department of Computer Science and Information Technology,
College of Engineering,
Abu Dhabi University, Abu Dhabi, United Arab Emirates
Email: osama.alomari@adu.ac.ae

Abstract: Carbon nanotubes (CNTs) have emerged in various applications due to their outstanding characteristics. The most common technique for producing CNTs with high yield and quality is known as chemical vapour deposition (CVD). However, manufacturers rely on conventional experimental studies to produce CNTs, which raise issues such as time, cost, and dealing with toxic materials. Alternatively, modelling and optimisation using metaheuristic algorithms are suggested to address these issues. This paper uses response surface methodology (RSM) for modelling work, while four metaheuristic algorithms are employed for optimisation. The regression and mathematical models, correlations, and significant CNTs process parameters are identified, analysed, and validated using RSM. The optimisation process and result are validated using different performance measure metrics and supported by other researchers. The CNTs yield and quality values improvement percentages in this paper are up to 36.45% compared to the referred original work.

Keywords: carbon nanotubes; CNTs; chemical vapour deposition; CVD; optimisation algorithms; response surface methodology; RSM.

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Biographical notes: Muath Jarrah received his master and PhD degrees in Computer Science from the Technical University of Malaysia, Melaka, in 2014 and 2018, respectively. He specialises in artificial intelligence and software engineering. His research interests include industrial computing, artificial intelligence, data science, machine learning, and modelling and optimisation algorithms. He has been awarded best doctoral thesis award in 2018 by Technical University of Malaysia. He has been working at different universities at different countries.

Zakaria N.M. Alqattan obtained his PhD from School of Computer Sciences and Mathematics in Universiti Sains Malaysia in 2017. He worked as a teaching fellow in National Advance IPV6 Centre for 2018–2019 at the same university. In 2020, he was a post-doctorate fellow at International Islamic University Malaysia (IIUM), Kuala Lumpur, Malaysia. He is now working as a Lecturer in the Department of Cyber Security and Cloud Computing Techniques Engineering, Northern Technical University, Mosul, Iraq. His research focuses on artificial intelligence, cyber security, intrusion detection and evolutionary computing including bioinformatics and optimisation.

Abdul Syukor Mohamad Jaya received his Bachelor's degree of BIT (Hons) in Industrial Computing from Universiti Kebangsaan Malaysia (UKM). He completed his Master in Mechanical Engineering (Advanced Manufacturing Technology) and PhD in Computer Science at Universiti Teknologi Malaysia (UTM). He is currently a Senior Lecturer in Department of Intelligent Computing and Analytics at Universiti Teknikal Malaysia Melaka (UTeM). His research and field of specialisation include applied artificial intelligence in manufacturing and robotics, and data analytics.

Sharif Naser Makhadmeh is an Assistant Professor at the University of Petra, Amman, Jordan. He received his BSc in Computer Science from Yarmouk University, Jordan, in 2013, and MSc in Information Technology from the Universiti Utara Malaysia (UUM), Malaysia, in 2015. He obtained his PhD degree in Artificial Intelligence from Universiti Sains Malaysia (USM), Malaysia, in 2020. His research interests include optimisation algorithms, artificial intelligence, engineering and scheduling problem, and smart home.

Ahmed Ismail Abu-Khadrah received his PhD in Computer Engineering and Communications from Technical University of Malaysia Melaka in 2017. He is currently at the Faculty of Computing and Informatics in the Saudi Electronic University. His research interests include wireless network protocols, networking, communications, wireless mathematical model, and multimedia service over the networks.

Ibrahim Aljarrah received his Bachelor's degree in Computer Science from Al-Balqa Applied University in 2018. He received his Master's degree in Data Science at Jordan University of Science and Technology in 2022. His research interests include data science, machine learning, deep learning, data analysis, and text mining.

Osama Ahmad Alomari is an Assistant Professor at the Abu Dhabi University, Abu Dhabi, United Arab Emirates. He received his BSc in Computer Science from Al Al-bayt University, Jordan, in 2005, and MSc in Computer Science from Universiti Kebangsaan Malaysia (UKM), Malaysia, in 2012. He obtained his PhD degree in Artificial Intelligence from Universiti Sains Malaysia (USM), Malaysia, in 2018. His research interests include artificial intelligence, data science, optimisation, feature selection, and biomedical data analysis.

1 Introduction

In our modern life, nanotechnology is an advanced and latest technology rising worldwide with endless research and applications of modern manufacturing technology (Anzar et al., 2020); it has revolutionised many areas and become the focus of many different industrial fields (Newberry, 2020). It is expected to be extensively used in multidisciplinary fields and future applications to make our life more sustainable, safer, and simpler (Balghusoon and Mahfoudh, 2020; Jadhav and Jadhav, 2018).

Carbon nanotubes (CNTs) have been the most exploited nanomaterials in the field of nanotechnology for over 25 years (Sett and Bhattacharyya, 2020; Miyashiro et al., 2020). Scientists and researchers have developed an immense interest in CNTs due to their excellent physical and chemical properties, such as ultra-light weight, high tensile strength, high thermal and chemical stability, and special electronic structures (Sari et al., 2018). With these outstanding properties, CNTs have emerged in various applications as excellent materials in medicine, material science, and electronics, such as sensors, fuel cells, televisions, and computers (Jadhav and Jadhav, 2018). CNTs are ideal electrical conductors because of their lightweight, high conductivity, current-carrying capability, strength, and thermal conductivity. CNTs are employed in various consumer applications because of their exceptional physical and chemical capabilities, such as in device modelling, energy storage, automobiles, water purification, sports goods, coatings, thin-film electronics, electromagnetic shields, and actuators. CNTs are promising drug delivery vehicles for anticancer medicines of both the biomolecule and small molecule varieties. Functionalising CNTs correctly improves their biological performance and makes them less hazardous, allowing them to be used in anticancer drug delivery.

However, the manufacturing process of CNTs-based products is still complicated and expensive, which limits the commercialisation and application of expensive products and luxury (Miyashiro et al., 2020). For this reason, engineers and manufacturers focus progressively on large-scale mass production to commercialise this fine technology (Sahu et al., 2018). Flexible CNT fabric and its prospective uses in technical and smart manufacturing are discussed in this research, along with the production technique for customised materials. Producing CNTs may be done in one of three major ways: arc discharge, laser ablation of graphite, and chemical vapour deposition (CVD).

Therefore, low-cost fabrication of CNTs with high percentage yield and quality is highly demanded to improve, strengthen, and produce robust CNTs-based products; it is still a challenge and the bottleneck problem for industrial production in large-scale applications (Sahu et al., 2018; Isoldi et al., 2020; Zhou et al., 2017). The lack of solubility, circulation half-life of 3–3.5 hours, non-biodegradability, immunogenicity, and biocompatibility are challenges of CNTs. There are several potential technical uses for CNTs, including their potential use as biocompatible modules for delivering bioactive. CNT uptake properties make them appropriate for biomedical applications, particularly gene and drug delivery. However, several issues with CNTs prevent them from being used in nano-medical applications. These include their tendency to bundle together, hydrophobicity, insolubility, presence of contaminants, and huge surface area (which leads to protein opsonisation).

CVD is the most common technique to produce satisfactory CNTs with property control and a high degree of structure. In fact, it is a versatile and economical technique for potential industrial applications. Due to its low cost, ease of use, and ability to manipulate CNTs on a wide scale, CVD has emerged as the preferred approach in recent years. CVD coatings are generally better in many applications where the materials and tolerances allow them. The problems of slide friction wear-out and galling are particularly common in metal-forming applications. CVD is superior to PVD because it forms a metallurgical and diffusion-type link between the coating and the substrate. However, the 1,925°F and 1,875°F processing temperatures used in CVD coating raise certain safety concerns. Due to tolerance issues, the use of CVD may be limited by the high processing temperature (Jarrah et al., 2016; Isoldi et al., 2020). This study evaluates the efficiency of CVD in relation to PVD based on the proposed metaheuristic algorithms, such as GA, PSO, BA, and ABC for optimum results.

Manufacturers rely on experimental studies that raise many issues regarding the experimental production process, such as time, cost and dealing with hazardous and toxic materials. This conventional method also does not guarantee to obtain the optimal CNTs production process parameters combination (Jarrah et al., 2019; Yusoff et al., 2019). However, reducing the trial and error experiments is the key solution for this issue. Thus, there is a need to maximise CNTs yield and quality by conducting fewer experimental trials, which could be done by modelling and optimising works (Fauzi et al., 2017; Chehrazi et al., 2017). However, the notion of optimising CVD parameters to study their impact on CNTs development is not yet completely formed (Jaya et al., 2015; Mohammed et al., 2017). The electrical conductivity of pure CNT may reach values as high as 10^6 to 10^7 S/m, whereas that of pure graphene can reaches as high as 10^8 S/m. These values are close to silver (6.30×10^7 S/m) and copper (5.96×10^7 S/m), two of the greatest metal conductors.

Modelling helps in analysis, predicting, and designing nanomaterials, in addition to finding significant parameters that influence CNTs characteristics (Chehrazi et al., 2017). RSM is the most mentioned technique of DOE, which is efficiently used for modelling purposes in manufacturing, regardless matter how complicated the relationships are (Jaya et al., 2015). RSM has been performed in Mohammed et al. (2017) to model CNT's crystallinity as an indicator of CNT's quality. CNTs quality indicated by ID/IG responses was studied using RSM, where experimental runs were designed and analysed by analysis of variance (ANOVA) a purposeful investigation of the connection input parameters and output responses. The result showed that RSM performs a high accuracy modelling work. Ghazal et al. (2022) synthesised CNTs using CVD based on the experimental design of RSM, where 20 experimental runs were suggested by RSM to investigate three process parameters and their effect on the CNTs' yield for low-cost and large-scale production. The study demonstrated the significant of using RSM in CNTs manufacturing. RSM is also used for optimisation purposes. The expanding electronic devices employ transistors and conductive layers based on the suggested response surface methodology (RSM)-based CNTs. Because of this, CNTs may be used as a substitute for Indium tin oxide transparent conductors in specific environments.

On the other hand, in CNTs production, optimisation can be defined as the approximation of the optimal process conditions; it could become the choice for the experimentation process to improve CNTs yield and quality. Optimisation using metaheuristics is a subset of artificial intelligence (AI). AI is a promising technology to solve current and future challenges (Ghazal et al., 2022, 2021; Ahmed et al., 2022). Metaheuristics may deal with quasi, quasi, or chaotic goals (Zavala et al., 2014). It has been proven that metaheuristics are efficient and perform superior in dealing with optimisation problems (Eshtay et al., 2019; Hamadneh, 2018). Bat algorithm (BA), particle swarm optimisation (PSO), artificial bee colony (ABC), and genetic algorithms (GAs) are well-known and widely used algorithms to solve optimisation problems in nanotechnology and similar manufacturing domains; they have shown better performance compared to the approaches at the cutting edge of optimisation science that may be used to a large variety of different fields of use (Al Nuaimi and Abdullah, 2017; Jarrah, 2018).

Owais et al. (2020) successfully implemented PSO in continuum approximation for CNTs. PSO could optimise the near minimum configurations on the surface of potential energy. Moreover, BA was applied in two case studies for CNTs growing in Jarrah et al. (2019); the optimal solution which maximises the CNTs yield% was obtained by BA. However, to our knowledge, lacking application of metaheuristic algorithms is obvious in optimising CNTs production using CVD. RSM is the most used technique intended to optimise CNTs production using CVD. This gives the motivation to apply and investigate the performance of metaheuristics for this particular field in this paper.

Graphene sheets are wrapped up neatly to create hollow cylinders, which are essentially what single-walled CNTs represent; in contrast to SWCNTs, multi-walled carbon nanotubes (MWCNT) consist of several layers of graphene rolled up uniformly into a tube shape. In addition, because of their exceedingly small diameter and large surface area, SWCNTs tend to exist in stiff, rope-like bundles, where the van der Waals forces between the particles are amplified. MWCNTs, in contrast, may be found in various shapes, including agglomerates, curls, and needles (Allaedini et al., 2016).

In this paper, five case studies are selected from the literature to maximise CNTs yield and quality using the CVD process; the case studies are selected from previous works in Bajad et al. (2015), Chai et al. (2011) and Lee et al. (2010). CNTs quality is represented by ID/IG, which is the D-band to G-band peak intensity. The ID/IG is calculated from a Raman Spectroscopy, where ID refers to the D-band, which indicates the disorder or the degree of defects that adhere to the structure of CNTs, including amorphous carbons (non-graphitic), while the G-band describes the well-ordered sp2 carbons in a graphite-like structure or the crystallisation level of the CNTs in their as-produced state. The following procedures were taken to acquire the multiple response optimisation process parameters and investigate the influence of many factors on the outcome: To provide the best possible conditions, numerical optimisation is employed to establish the objectives for each response. Minimising degradation and increasing the hardness of metal matrix composites is the focus here, and this study has chosen to concentrate on the minimum values for the responses and the ranges of the components. The superimposed graph is created by setting minimum and maximum limits for each response using graphical optimisation, therefore identifying a feasible range of values.

For optimisation, the mathematical models or regression obtained by RSM for the primary three examples, see the source material. However, the objective functions of the other two neither introduction of research papers could be found in the primary source. Therefore, they are modelled and analysed in this paper using RSM to find their objective functions. Objective functions are used in optimisation algorithms to find the optimal output. Optimisation algorithms entail adjusting the model parameters for a given set of inputs. The best may be evaluated in different contexts regarding time, resources, accuracy, etc. The prediction algorithms use the known model parameters to approximate the system's output behaviour at a given input state. The modelling work in all case studies is performed using RSM. In the optimisation phase, GA, PSO, BA, and ABC are applied to the case studies to carry out the optimum yield and quality of CNTs, and their corresponding solutions. Precise and detailed settings of algorithms' parameters are covered in the tuning process to ensure that the best parameters are selected for the best result while covering most of the algorithms' search space. The tuning process and setting points from this paper could be used by other researchers to improve the performance of the algorithms and the solution to real-world problems. In addition, the finding of this paper suggests using metaheuristics as an alternative technique to RSM for optimisation. Besides, engineers and manufacturers can use the reported output from the modelling work and optimisation techniques in real-world production to maximise CNTs' yield and quality. Further, it could be used as a trusted guideline to control and customise CVD process parameters to estimate the best CNTs characteristics.

Here's how the remainder of the paper is laid out: the study's approach is described in Section 2 of this paper. The description of the case studies is presented in Section 3, including their process conditions and parameters. In Sections 4 and 5, the modelling work using RSM is introduced. After that, the optimisation process using metaheuristics is presented in Section 6. Sections 7, 8, and 9 discuss result analysis and validation and its effect on the actual synthesis of CNTs. Finally, Section 10 concludes the paper.

2 Methodology

The research methodology primarily focuses on combining regression and optimisation. Regression is based on RSM, while optimisation is based on selecting metaheuristic algorithms to be adapted to the case studies of CNTs growing using the CVD process. Single-and MWCNT may be synthesised using CVD by catalytically decomposing a carbon precursor (such as CO, hydrocarbons, or alcohol) on a nanostructured transition metal catalyst such as Co, Ni, or Fe. Temperatures between 600°C and 1,000°C are common for CVD. The optimisation aims to find the optimal values of CNTs yield and quality with their corresponding input parameters. The overview of this study is illustrated in Figure 1, which comprises three main phases; data collection, modelling, and optimisation. Firstly, the selected data are described and chosen to represent the case studies in this paper. Then, the modelling work defines the mathematical regression equations to be used as objective functions in the last phase of the optimisation process using state-of-the-art algorithms, namely BA, GA, PSO, and ABC. Finally, the optimisation process is detailed with the parameters' tuning strategy. This study collects samples from the dataset of the UCI machine learning repository (https://archive.ics.uci.edu/ml/datasets/Carbon+Nanotubes) for CNT calculation parameters.

Data collection includes data definition and description of the case studies. In this paper, five case studies are presented, comprising four separate instances to optimise CNTs produce results, and there is at least one example of optimising CNTs quality, represented by the ID/IG ratio. The intensity ratio of the Raman G-band and D-band is often utilised to measure the thickness of structural defects in CNTs, providing a comparative measure for the structural quality of a sample. If both bands are similar in intensity, the thickness of structural defects is assumed to be high. Conversely, a lower ratio indicates fewer defects and greater structural quality. The case studies are selected to cover a wide range of process parameters, the most commonly studied in CNTs growing using the CVD process. By covering a wide range of process parameters, the yield and quality of CNTs could be optimised in different conditions, and the tested algorithms and their performance will be investigated with different search spaces, such as simple with two input parameters and more complex with four input parameters. Central composite design (CCD) is a statistical technique based on a multivariate nonlinear model that has been extensively used to optimise the process variables of biosorption. It has also been used to determine the proper experiments' regression model calculations and operating conditions. Box-Behnken designs (BBDs), like CCDs, can accommodate a whole quadratic model since they are response surface designs. In addition, BBDs have fewer layers per factor than other centralised composites. So, they are appealing when the elements are quantitative, but the range of possible values is limited.

Section 3 and Table 1 describe the CNTs' growing process parameters in different case studies. Summary descriptions of the featured research papers are provided in the next section.

3 The description of the case studies

This section highlights a short explanation of the chosen research papers for CNTs growing as follows:

3.1 Case study 1: 'synthesis of CNTs by methane decomposition over Co-Mo/Al_2O_3 process study and optimisation using RSM'

The CNTs growing experiment by Chai et al. (2011) to measure carbon yield was considered in this study. CVD was used for the growing process. The CNTs were synthesised over Co-Mo/Al_2O_3 catalyst by methane decomposition. The carbon yield, the percentage of gas conversions was approximated based on data gathered from a real-time gas chromatograph (GC) examination (Hewlett-Packard Series 6890, USA). This dataset aimed to use RSM to maximise the CNTs yield using three process parameters: catalytic mass, precipitable water of gas, and catalyst loading. Therefore, the RSM approach with a three-level factorial design with the three process conditions was adopted in this case study. A RSM was used to determine the optimal process parameters for optimising CNT yield while minimising amorphous carbon structure. The best conditions for running the CVD reactor that result in the highest production and purity of CNTs were identified using response surface methods. This study investigated the effects of catalyst/condensate-gas weight ratio (5–10 wt%), synthesis duration (30–120 min), and temperature (700°C–1,000°C) on CNT synthesis. The catalyst/condensate gas mass ratio of 5%, the synthesis duration of 112 minutes, and the temperature of 1,000 degrees Celsius were determined to be optimal for CNTs-synthesis. Raman spectroscopy shows superior CNT quality under these conditions, with ID/IG ratios in the high range.

3.2 Case study 2: 'synthesis and characterisation of CNTs using polypropylene waste as precursor'

The CNTs growing experiment by Bajad et al. (2015) to measure carbon yield was considered in this case study. CVD was used for the growing process. The hydrogen precursors from PP container trash were studied using a Ni/Mo/MgO catalyst blend by combustion. This case study considers four growing conditions for CNTs growing process. They are the factors of process duration, catalytic dose, polymeric mass, and heat. This dataset aimed to use RSM to maximise the CNTs yield by studying the effect of the four parameters process. This case study adopted the RSM approach with BBD with the four process conditions. Experimental design with three levels of the CNTs growing process condition is given in Table 1. A CVD machine was used for the growing experiments. The required surface quality is crucial to the machining process. Most mechanical products have technical criteria of surface roughness or the intended surface index of product quality. In addition to being a crucial aspect in the machining process, the surface roughness (quality) value is a useful measurement to evaluate the finished product's quality. Machine parameters, including cutting speed, feed rate, depth of cut, and cutting tool entering angle, all have a role in the surface roughness of a machined component.

3.3 Case study 3: 'yield optimisation of nanocarbons prepared via chemical vapour decomposition of carbon dioxide using surface methodology'

The CNTs growing experiment by Allaedini et al. (2016) to measure carbon yield was considered in this case study. CVD was used for the growing process. CO_2 was considered a carbon source, and the MgO substrate-supported Ge nanoparticles catalyst (Ge/MgO) was employed. This work-study examines the influence of experimental parameters on the production of CNTs employing CVD by the direct breakdown of CO_2 in the absence of nanocomposites from the chemical family. In this case study, three growing conditions are considered for the CNTs growing process the CO_2 rate, the heat of the process, and the amount of catalyst used. This study aims to optimise CNT production and investigate how environmental factors influence CNT growth.

The experimental design for the CNTs growing process is given in Table 1. The two levels of the growing condition of experimental design are –1 (low) and +1 (high). A CVD machine was used for the growing experiments. The SEM and TEM photos revealed the existence of CNTs, while electricity cross spectrometry (EDS) (Zeiss SUPRA55) data were utilised to assess the quantity of carbon deposition by examining the mass % of the components contained in the produced powder. It should be noted that the catalyst loading was eliminated from the regression model in the original work (Allaedini et al., 2016) due to its insignificant effect on CNTs yield.

3.4 Case study 4 and 5: 'optimisation of CNTs synthesis via methane decomposition over alumina-based catalyst'

The CNTs growing experiment by Lee et al. (2010) to measure carbon yield and quality was considered in generating case studies 4 and 5. The two different output responses with the same input parameters and values were used; the first is carbon yield representing case study 4, while the second is ID/IG ratio, which indicates the quality of the CNTs and represents case study 5. The aim is to maximise the CNTs yield in case study 4 and to minimise the ID/IG ratio in case study 5. Experimental design with a tiered BBD concentration-wise, having a single focus for the process condition of CNTs growing, is given in Table 1. The RSM approach with three-level BBD and the three process conditions were adopted. The selected datasets of the case studies are introduced in the original works in Allaedini et al. (2016), Bajad et al. (2015), Chai et al. (2011) and Lee et al. (2010).

CVD was used for the growing process. The CNTs were synthesised by breaking down gas using CCVD to a catalytic built-on oxide ($CoO\text{-}MoO/Al_2O_3$). Carbon deposition percentage was determined using the loss of materials between 300 and 700C and was measured using an analysis [TGA analysers (TGA); TA Equipment, SDT Q600]. For ID/IG, an inVia Raman Microscope (Renishaw) was employed to record Raman spectrums using a 532 nm laser for illumination, with a range of wavelengths of 100–3,200 cm^{-1}.

In these latter case studies, three growing conditions are considered for the CNTs growing process. They are the aspects of the process, such as heat, time, and metallic load.

4 Modelling and optimisation

To fulfil the optimisation process, the objective function of each case study is mathematically represented to be used by the optimisation algorithms. In this paper, RSM was conducted for modelling purposes to find correlation and regression to generate the objective functions of the selected case studies. However, among the five case studies, only three cases were modelled in the original works and represented by their objective functions that can be used in optimisation algorithms to search for optimal CNTs yield and quality values. In other words, the models in the last two cases were developed using RSM for coded data, while the models for actual data were not mentioned in the referred works, meaning that the reported models in the referred works cannot represent the objective functions for metaheuristic algorithms for optimisation purposes. Therefore, a modelling work of these two case studies is developed in this paper to be used by metaheuristic searching algorithms to find the optimal yield and ID/IG values.

Therefore, RSM modelling work and development in this paper is used for only two case studies: case study 4 and case study 5. Hence, the regression models in the authors' first three case studies resources are fully referred to and used to formulate the objective functions for the optimisation problems of GA, PSO, BA, and ABC for growing CNTs. Table 2 summarises the objective functions of all case studies. However, data-driven RSM modelling work for case study 4 and case study 5 is discussed in Section 5.

5 RSM modelling work

RSM could be defined as a sequential procedure and collection of statistical and mathematical techniques used for designing experiments to analyse, model, and optimise the process. The dependent variable, i.e., the output response variable influenced by the independent variables, i.e., process input parameters (Montgomery, 2017). The RSM methodology in this study is shown in Figure 2.

In the RSM-based model, to approximate the responses, a quantitative representation of independent process parameters is employed as follows in equation (1):

$$y = f\left(x_1, x_2, x_3, \dots x_n\right) \pm \varepsilon \tag{1}$$

As shown in equation (1), where y represents the reaction output or dependent variable, f is a functional reaction, x_1, x_2, x_3, …, x_n the gratis input the gratis ε is the incorrect placement. The approximation of f determines the suitability of RSM. The regression model in a second-order polynomial is used to obtain f, also called the quadratic model. The quadratic regression model of f is given in equation (2):

$$f = c_0 + \sum_{i=1}^{n} c_i x_i + \sum_{i=1}^{n} c_i x_i^2 + \sum_{i,j}^{n} c_{ij} x_i x_j + \varepsilon \tag{2}$$

As inferred from equation (2), the quadratic regression model is calculated, and where (c_0) is constant, (n) is the number of variables, and the three summations represent the linear, quadratic, and interaction terms, respectively, where c_i, c_{ii} and c_{ij} are their coefficients, respectively. (x_i) and (x_j) denote the explicative factors or independent variables, and (ε) is a standard error, also called statistical error or random experimental error (Khor et al., 2016).

Based on (2), it is expected that the principal component variables (i.e., reaction temperature, reaction duration, and metal loading) would have various effects on carbon yield and quality in CNTs synthesis factors, including exponential, exponential, and let's try. Thus, the regression models could be written as in equations (3) and (4):

$$\begin{aligned} y_{yield}(x) = {} & c_0 + c_1 x_1 + c_2 x_2 + c_3 x_3 + c_{11} x_1^2 + c_{22} x_2^2 + c_{33} x_3^2 \\ & + c_{12} x_1 x_2 + c_{13} x_1 x_3 + c_{23} x_2 x_3 + \varepsilon_{yield} \end{aligned} \tag{3}$$

$$\begin{aligned} y_{ID/IG}(x) = {} & \gamma_0 + \gamma_1 x_1 + \gamma_2 x_2 + \gamma_3 x_3 + \gamma_{11} x_1^2 + \gamma_{22} x_2^2 + \gamma_{33} x_3^2 \\ & + \gamma_{12} x_1 x_2 + \gamma_{13} x_1 x_3 + \gamma_{23} x_2 x_3 + \varepsilon_{ID/IG} \end{aligned} \tag{4}$$

where y_{yield} and $y_{ID/IG}$ represent the carbon yield and CNTs quality, respectively. c_0 and γ_0 are constants, while c's and γ's are the coefficients of the independent variables x's. The impacts of the cubic, nonlinear, and interaction factors may be calculated using these quadratic regression models matching these equations to the experiments. Thus, the entire factor space is investigated, and the region of optimality is located (Seo et al., 2017).

The modelling process includes developing CNTs yield and quality indicator (ID/IG) models concerning the CVD process. The growing process behaviour based on significant findings is also discussed in this paper. Besides, the interaction of the process parameters that affect CNTs yield and ID/IG are also identified. A mathematical equation for each model is developed to represent the relationship between the CVD process parameters and results in the form of input. Furthermore, the calculation tool is tested and proven accurate by correlation and t-test to investigate the similarities between the models' results and the experimental data.

5.1 *Estimating the impact of CVD variables on CNT output modelling (case study 4)*

Thirteen experimental runs and the result of the dataset for CNTs yield are given in Lee et al. (2010). The yield of CNTs varies from 170.8% to 404.3% with different input parameter values. In this study, the first step was determining what change was needed to begin the analysis. The study performed in Design Expert (V 8.0.0) shows that no conversion is required since the lowest to fulfil the demands of the 's getting is 2.37, and a voltage convert has a negligible impact in this region (< 3). The change must be made if the proportion is more than 10 (Jaya, 2013). Besides, model transformation terms can be determined by using diagnostic plots. In this study, Figure 3 represents three significant plots to be analysed for the developed yield model. The first plot is residual normality, where the studentised residual is analysed using a normal probability plot. The second plot is the studentised residual versus the predicted value to check the model's constant errors, and the third plot is the external plot of the studentised residual, which is analysed to look for outliers.

Errors follow a normal distribution, as seen by the normal curve in Figure 3. When the residuals cluster near a single direction, we may understand this phenomenon. As a result, no change in the answer is needed. In the meantime, the plot of expected response vs. residue for CNTs yields shows no clear pattern or out-of-the-ordinary creation of the data. There is also no discernible speaking pattern, and the storyline is dispersed in an unorganised fashion. Since this trend already exists without the reply conversion, this finding confirms that the latter is unnecessary. All the shown data for residue vs. cycle numbers for CNTs yield are from the eight red lines. Because of this, it is unnecessary to do research to determine the source of the anomalous results. The organisation and acceptable pattern shown in the graphs plots suggest that no modelling or reaction terms modification is necessary. Furthermore, the residual data in the plot is within an acceptable range; thus, there is no need to adjust the experimental data used to represent CNTs output inside the CVD input variables.

5.1.1 Quadratic formula for representing CNTs' yield in an RSM model

The model analysis is needed to determine the appropriate model representing the relationship between CNTs yield and CVD process parameters. For this purpose, the sequential model sum of squares (SMSS) analysis and model summary statistics are analysed.

The model selection criterion in SMSS was at the polynomial with a significant p-value ($p < 0.1$) and not aliased. Therefore, the source of 'quadratic vs. 2FI' is suggested in this study due to the higher order polynomial term and not aliased. Unfortunately, the experimental runs are insufficient to estimate all the terms for the models.

On the other hand, the result from the model summary statistics analysis suggests the source quadratic due to low standard deviation, R-squared is close to 1, and maximum adjusted R-squared, where the best result is found at max r value and anticipated R-squared, highest adjusted R-squared, and poor threshold deviation.

5.1.2 ANOVA of the quadratic model for CNYs yield

ANOVA is required to identify the key factors and relationships that affect CNTs output. The created model is not meaningful at a p-value of 0.1431 according to the first ANOVA study of the response surface quadratic polynomial to CNTs yield. RSM's first-order, second-order, and mixture models will each be subdivided and extracted in detail. In the CNT process, the advantages of RSM include learning a great deal through limited experiments on historical literature extraction circumstances for case study outcomes. In addition, exploring factors' primary and interactive impacts on the response may be facilitated via models and graphical information. When the p-value is below 0.1, it is considered statistically significant. If the probability value above the likelihood ratio (Prob. > F) is less than 0.05, then the model terms are statistically significant. In this scenario, we have three variables designated by the letters A, B, and C: the heating rate, the reaction time, and the amount of metal added. The only reaction temperature is a significant model term. Therefore, a model reduction may improve and achieve a significant model.

The manual elimination method has successfully improved the model's significance and hierarchy. In this method, the model reduction was implemented by removing the insignificant terms that affect the model. The details of model reduction and improvement are shown in Table 3. One superficial description could be that the poor CNT structural quality induced by growing CNT arrays at low temperatures hides the CNT yarn metallic behaviour. Despite a significant increase in the CNT structural quality by annealing treatment, our experiments have clearly observed a more robust semiconducting behaviour.

The significance of the model is shown by the F-value of 4.45. The likelihood of seeing a 'model F-Value' due to random noise is very low at 4.60%. Model terms are statistically significant since their p-value (Prob. > F) is 0.046, lower than 0.05. Values greater than 0.1 indicate the model terms are not significant. In this case, B and C are more significant with a p-value of 0.0188 and 0.0256, respectively, while the terms A, AC, BC, and C^2 have less significance on the model. Additionally, the developed model summary statistic indicates that the model has a low standard deviation of 47.29. Therefore, the R-squared value is close to 1. The predicted R-squared 'Pred. R-squared' one would imagine, the value of 0651 is not very near to the value of 0.6332 for the adjusted R-squared. This suggests that there may be an issue with the empirical observations. The sensor ratio is quantified by adeq. accuracy. Therefore, it's preferable to have a ratio higher than 4. With a model ratio of 6.395, sufficient evidence supports the conclusion.

5.1.3 Identification of key aspects that play a CNTs yield

Based on the signature model, due to their low p-values, terms B and C have a more substantial influence on the system than term A. 0.05. Also, the word of the cubic form of metal loading (C^2) is significant with a p-value < 0.1. The other terms have less effect on the model; the effect sequence order of terms is B > C > C^2 > BC > AC > A. The behaviour of the model terms will be compared, as shown in Figure 4, with other related works to support findings from the developed model in this paper. However, since the parameters have three alpha values that are difficult to represent in this study, the middle point of the third parameter is selected for fair analysis of the interaction terms.

5.1.4 Changes in the temperature and their relationship (A) and a heavy intake of metal (C) the moment of truth: reaction speed (B) of 2.5 hours

At B = 2.5 hours. Figure 4(a) shows that at a constant 10% of metal loading, an increase of temperature from 700°C to 800°C decreases the CNTs yield from 231.75% to 163.85%, respectively. However, the trend is reversed for higher metal loading at constant 40%, where the CNTs yield performance increases from 290.63% to 302.33% as temperature increases from 700°C to 800°C, respectively. Therefore, there is a strong interaction between the two process parameters. This finding is supported in Figure 4(b) in the 3D surface graph and Figure 4(c) in the contour graph. At a constant reaction time of 2.5 hours and low metal loading of 10%, the CNTs yield decreases by increasing the temperature. Conversely, the CNTs yield increases by decreasing the temperature at low metal loading of 10%. In addition, at higher metal loading greater than 37%, the CNTs yield increases by increasing temperature. Conversely, at higher metal loading greater than 37%, the CNTs yield decreases by decreasing temperature. From Figure 4(b), it is clearly indicated that the best CNTs yield of 319.7% could be obtained at a temperature around 700°C and 29% of 2.5-hour metals load response. This interaction phenomenon between temperature and metal loading is a significant finding and could be used to adjust and control the CVD process parameters for growing higher and optimum CNTs yield.

It is worth noticing here that this result is in agreement with the experimental work in Lee et al. (2013), where they synthesised CNTs using the same conditions over cobalt oxide-molybdenum oxide/alumina (CoO-MoO/Al_2O_3) catalyst by methane decomposition at 700°C, two hours and different metal loading% of 10%, 20%, 30%, and 40%. The authors found that increased metal loading from 10% to 30% makes the crystallite amount of CoO more favourable for more CNTs growth, resulting in increasing CNTs yield. Meanwhile, at 40% of metal loading, the clusters of CoO were too large at 700°C and exceeded the favourable range of CNTs synthesis, resulting in decreasing CNTs yield at 40% of metal loading compared to 30%. Thus, the optimal CNTs yield at 700°C for two hours was found at 30% of metal loading. In addition, the CNTs diameter was found to be broader at these syntheses conditions by increasing the metal loading%. Figure 5 illustrates the trend of the carbon yield at different metal loading% at 700°C for two hours (a) and the transmission electron microscopy (TEM) image of grown CNTs for 30% metal loading at 700°C for two hours (b).

5.1.5 Impact of increasing concentration on response time (BC) the heat of 750°C

The optimum result at 750°C was close to 700°C at 388.39% and 388.7%, respectively. This result indicates that the reaction temperature (A) of 750°C and at 700°C has the same impact on the CNTs yield.

Based on the CNTs yield modelling work, since the developed model has been analysed and proven to be significant, a quadratic polynomial equation that represents the relationship between CNTs syntheses process parameters and the response of carbon yield could be written as in equation (5). A quadratic polynomial equation is established to forecast the responses as a role of independent parameters, including their quadratic interactions and squared terms (Mahmoodi et al., 2020).

$$\begin{aligned} CNTs\ yield\% = {} & +787.00639 - 0.94433 R_T - 4.32500 R_D - 8.48194 M_L \\ & +0.026533 R_T M_L + 1.59667 R_D M_L - 0.24241 M_L\hat{\ }2 \end{aligned} \tag{5}$$

where R_T is reaction temperature, R_D is reaction duration and M_L is metal loading.

5.2 Modelling of ID/IG ratio for CNTS concerning CVD process parameters (case study 4)

Thirteen experimental runs and the result of the dataset for CNTs quality are given in Lee et al. (2010). The data indicates that the ID/IG ratio varies from 0.455 to 1.241 with different input parameter values. Since the lowest to fulfil the demands of the s' set is 2.727, which has minimal influence on voltage conversion at less than 3, the study suggests that no conversion is necessary. No reply modification is necessary since the mistakes follow a normal likelihood function, as shown in Figure 6(a). On the other hand, Figure 6 depicts the ID/IG projected response vs. residual plot (b). This chart shows that the data exhibits no peculiar trends or patterns. There is no discernible megaphone pattern, and the story is dispersed in a disorderly fashion. Since this pattern already exists without the reply conversion, this finding confirms that the latter is unnecessary. The structure and acceptable pattern shown in the graph plots suggest that no modelling or reaction term modification is necessary. No adjustments to the experiments are necessary, either, since the show's residual data are within the expected range needed in representing ID/IG in CVD process parameters.

5.2.1 Determination of polynomial equation to represent RSM model of ID/IG

SMSS shows that 'linear vs. mean', '2FI versus linear', and 'quadratic versus 2FI' have no significant p-values at $P > 0.1$. In addition, 'cubic vs. quadratic' is aliased. However, the source of 'quadratic vs. 2FI' is suggested in this study due to the higher order polynomial term and not aliased. Unfortunately, the experimental runs are insufficient to estimate all the terms for the models.

On the other hand, the standard error, R-square, r Value, and forecasted R-square are all analysed in the model descriptive statistic. When the standard error is small, the R-squared value is close to 1, and the corrected R-squared value and the projected R-squared value are both close to their maximum, getting the best results. The source linear is suggested due to low standard deviation, R-squared is close to 1, and maximum adjusted R-squared. Therefore, the suggested linear model is analysed and modified based on the analysis to represent the relationship between ID/IG response and its process variables.

5.2.2 ANOVA evaluating the polynomial models for ID/IG

The initial ANOVA modelling on a linearised surface to ID/IG showed that only the C factor in the linear term is significant. However, for optimisation, the model was modified by adding linear and quadratic terms to be significant and to fit the experimental data to be valid for optimising all studied process parameters. The manual elimination method has successfully improved the model's significance and hierarchy. As indicated in Table 4. The developed quadratic model or modified linear is significant with a 0.0447 p-value. Using the manual method, the model was improved by adding some terms. The new model has terms A, B, C, AB, BC, A^2, C^2, and A^2B. Meanwhile, all model terms are significant at a P-value < 0.1, except A quadratic term, which showed less effect with a P-value close to 0.1.

The model is statistically significant, with an F-value of 6.45. It is quite unlikely that a 'model F-value' would emerge by chance alone; the probability is just 4.47%. The regression coefficients are statistically significant since the p-value is 0.0447 (Prob. > F). If the probability value above the likelihood function (Prob. > F) is less than 0.0500, then the model terms are statistically significant. The model terms with the greatest impact in this example are B, AB, and A2B (P0.0439), whereas the variables with the least impact, are A, C, BC, A2, and C2.

Additionally, the developed model summary statistic indicates that the model has a low standard deviation of 0.095, and the R-squared value is close to 1. Compared to the 'adj. R-squared' value of 0.7843, the 'pred. R-squared' value of 0.0481 is rather off. This suggests that there may be an issue with the experimental data. The 'adeq. precision' metric also accounts for the signal-to-noise ratio. It's preferable to have a ratio higher than 4. In addition to the diagnostic plots analysed in Figure 6, the model ratio of 9.478 suggests an appropriate signal. As a result, the design space may be explored with the help of this model. This research examines the most important parameters with a P-value < 0.1 that influence the ID/IG value are presented in the next sections.

5.2.3 Length of reactions relating to temperature (AB) at 25% of a heavy intake of metal (C)

As shown in Figure 7(a), at a constant time of 1 hour, an increase of temperature from 700°C to ≈763°C decreases the ID/IG from 0.77 to 0.61, respectively. Next, the ID/IG increases as temperature increases to reach 0.669 at 800°C. However, the trend is reversed for a higher time at constant 4 hours; the ID/IG increases from 0.63 to 0.87 as temperature increases from 700°C to ≈745°C, respectively. Then, the ID/IG decreases as temperature increases to reach the lowest value of 0.526 at 800°C. Figure 7 shows the Behaviour of ID/IG in terms of how they interact with AB at C = 25%, interplay, 3D surface, and contour. In the CNTs sample, the relative intensity of the D band concerning the G band can be utilised to measure the concentration of defects. When the ID/IG ratio is small, the CNTs with graphite structures generate more than defect structures.

5.2.4 Way metals loading interacts with response time (BC) at 750°C

The effect of reaction duration is less significant where metal loading is low. Therefore, at low metal loading (10%), the effect of reaction duration can be reduced where there is no obvious effect on the ID/IG value. Also, at high metal loading of 40%, there exists a general increase in ID/IG as reaction duration increases, where the ID/IG increases from 0.8 to 1.27 at reaction duration from 1 hour to 4 hours, respectively. Figure 8 illustrates this finding and shows the value of ID/IG at the BC level of 10% for 1 hour is better than at 40% of metal loading.

This result is in agreement with experimental work in Lee et al. (2010) where they synthesised CNTs using the same catalyst and carbon source, which are CoO-MoO/Al_2O_3 catalyst by methane decomposition at closed parameters' values at 762°C, 2.3 hours, and 27% of metal loading% as shown in Figure 9. The authors found that low ID/IG ratio of synthesised CNTs producible at high reaction temperature and low metal loading. Two significant peaks are shown for the grown CNTs, representing the D-band and G-band at 1,359 and 1,584 cm^{-1}, respectively. The ID/IG ratio value for this case was 0.595.

Based on the ID/IG modelling work, and since the developed model has been analysed and proven to be significant, a quadratic polynomial equation that represents the relationship between CNTs syntheses process parameters using CVD and the response of ID/IG could be written as in equation (6):

$$\begin{aligned} ID/IG = {} & +54.27917 - 0.14106 R_T - 29.23033 R_D - 0.032843 M_L \\ & +0.077813 R_T R_D + 5.06667(E-003) R_D M_L \\ & +9.33095(E-005) R_T^2 + 6.16032(E-004) M_L^2 \\ & -5.18667(E-005) R_T^2 R_D \end{aligned} \tag{6}$$

where R_T is reaction temperature, R_D is reaction duration and M_L is metal loading.

5.3 Validation of CNTs growing characteristics for the developed models

The developed RSM model validation is investigated using a t-test on data pairs. Calculating correlation is to monitor the significant relationship between the developed RSM models with the actual experimental data. A paired sample test is a statistical method used to compare the significant difference between two results (Jaya, 2013). In this case, predictions from different models can be compared using the t-test developed RSM models with the actual experimental data.

Table 5 presents the correlation value of the experimental data and an expected output of 0.904% in CNTs. This strong connection suggests that the quantities anticipated by CNTs by the RSM model are almost similar to CNTs yield from the experimental data. Meanwhile, Table 6 shows the difference of means for the CNTs yield from the experimental and RSM prediction model is 0.00258, which falls between –20.205 and 20.2101 with a 95% confidence interval. The calculated t value is 0, lower than 2.179 of the critical t value from the standard table (the cumulative distribution function table for the t-distribution) when df equals 12. Further, the test's significance level is 1, significantly higher than the threshold of 0.025 (significant of 2-tailed). The median CNTs yield predicted under the RSM model is similar to the average of experimental CNTs yield.

On the other hand, related to the ID/IG response, Table 7 shows that the correlation value of the experimental data and the predicted ID/IG response is 0.963. This high correlation value indicates that the predicted values of ID/IG by the RSM model are almost similar to ID/IG values from the experimental data. Meanwhile, Table 8 shows the difference of means for the ID/IG from the experimental and RSM prediction model is 0.0043, which falls between –0.0288 and 0.03748 with a 95% confidence interval. The calculated t-value is 0.285, which is lower than 2.179 of the critical t-value from the standard table (the table of cumulative distribution function for the t-distribution) 12 df = moderate confidence interval. Further, the test's p-value of significance, at 0.781, is higher than the significance level of 0.025 (significant of 2-tailed). Given that the averages of the prediction system with the testing results are not statistically different, one can conclude that the RSM model's averaged ID/IG is quite similar to the average of experimental ID/IG.

Moreover, the plots in Figures 10(a) and 10(c) show the output distribution and pattern matching of CNTs yield and quality results of the RSM models versus the experiments, respectively. The plots show that the values of the experiments and RSM-predicted models distribute around the mid-line. Besides, Figures 10(b) and 10(d) show that the line patterns are almost similar for the experiments and RSM predicted models. This implies that the predicted result from the two developed models of RSM is almost similar to the experimental data. Thus, it is proven that the developed models using the RSM can predict the yield and ID/IG responses of CNTs with less error. In conclusion, with a high prediction accuracy, the developed RSM models can predict the yield and ID/IG ratio in the CVD technique for the CNTs growing process.

5.4 Impact of real-world CNT applications on CVD-grown CNTs

In this study, for a high yield of CNTs growing at 391.64%, RSM found the optimum parameters at the highest levels of temperature at 800°C, duration of 4 hours, and 40% of metal loading. These optimal parameters obtained better yield due to the efficient decomposition of methane with high thermal stability (Lee et al., 2010). Meanwhile, methane decomposition is deficient at low temperatures (Dai, 2002). Moreover, the yield is increased at a long duration of four hours and high metal loading of 40% because of increasing active catalyst sites, which leads to more room for CNTs to expand on the substrate (Lee et al., 2010). Another reason behind the high yield in this study could be that hydrogen facilitated the synthesis of CNTs (Lee et al., 2012).

On the other hand, for optimising the ID/IG ratio of CNTs, the optimal values of two parameters settings were found to be the same as in the CNTs output at 800°C and four hours, indicating that increasing scores of both reactions and duration are preferable for both CNTs production and quality low ID/IG ratio is grown at a high reaction temperature (Niu and Fang, 2008). In addition, better CNTs quality with a lower ratio of ID/IG can be synthesised at a lower value of metal loading of 10 with a high reaction temperature and long duration (Lee et al., 2010). The CNTs morphology and diameter would be influenced by reaction time (Sivakumar et al., 2010).

In hydrocarbon CCVD, hydrogen can be treated as a reactive gas, which renders a reducing environment for the catalyst particles in the reaction chamber. Hydrogen also helps the catalyst activity since it has been regarded as a promoter for the catalyst (Xiong et al., 2005). In addition, it was proven that hydrogen selectively etches the amorphous carbon, thus preventing the active sites from poisoning by the deposition of amorphous carbon and undesirable materials (Lee et al., 2012). This revealed that the hydrogen from methane hydrocarbon plays a significant role in enhancing the CNTs quality with low ID/IG in this study.

Also, for this case study, it was reported that the metal support interaction (MSI) and supported material, i.e., CoO-MoO and Al_2O_3 play a significant function in the enzymatic breakdown of methanol into hydrocarbons and oxygen. Because of the increased reducing temperatures and the creation of metal-support species that are hard to process and even recalcitrant, high MSI slows the gradual decrease of reactive metal oxide. In addition, high MSI prevents metal particles from clumping together on the support surface, allowing for further even distribution of the metals throughout the material (Taust, 1987; Awadallah et al., 2014).

In addition, both morphology and yield of the as-produced CNTs are drastically influenced by combining transition metals with Mo in bimetallic catalysts (Awadallah et al., 2012). The transition metals catalysts, i.e., Co in nanoparticles form, are very effective because, at a high temperature, carbon has a high diffusion rate and high solubility in these metals. This could support the result of the optimum yield and quality from RSM at a high-temperature value. Furthermore, it was postulated that the synthesis of CNTs is assisted by Mo species in two ways:

1 Mo species help the dispersion and avoid the sintering of cobalt metallic particles, confirming the active sites.

2 They help in the aromatisation of the hydrocarbon source and generate intermediate aromatic species which act as growing units for CNTs production (Shah and Tali, 2016).

In conclusion, the RSM modelling result is appropriate for estimating CNTs yield and quality. Therefore, RSM is highly recommended for this case and similar cases.

6 Improving the quantity and quality of CNTs

In this section, the increasing production and improving grade (ID/IG ratios) of CNTs is done using four common metaheuristic algorithms, including GA, PSO, BA, and ABC. In this process, precise and detailed settings of algorithms' parameters are covered in the tuning process to ensure that the best parameters are selected for the best result. Besides, the topic of discussion comprises detailed results and analysis of the optimisation process, in addition to the performance of the applied algorithms. Finally, the optimisation result of CNTs and their effect in real experiments synthesis is discussed and supported by previously published works.

The selected five case studies were described in Section 3. It should be noted that the experimental data of the case studies are collected in different process conditions, i.e., different input parameters and a different number of these parameters, different substrates, and different catalysts. Since this study aims to investigate both the yield and quality of CNTs, the proposed optimisation algorithms should be adapted to tackle different case studies in different conditions. Therefore, each algorithm is coded particularly for each case study separately, and the best algorithm for each case study will be considered to represent the certain investigated case study.

6.1 Experimental design

This section includes setting default parameters of the used algorithms in this paper, solution representation, and performance measurement metrics.

6.2 Optimisation constraints of the case studies

The objective functions of the optimisation algorithms for carbon yield and quality for the case studies can be written in Table 9.

All input parameters in the objective functions are subjected to the limitation constraints of $input_{\min} \le input \le input_{\max}$ as indicated in Table 1.

6.3 Gains in durability and productivity of CNTs

In all presented instance scenarios, the optimal results were obtained in the original works by using RSM. Therefore, to evaluate the proposed algorithms' efficiency, the tested algorithms' optimum results should be close to or better than the introduced optimal output in the original works. Table 10 displays the optimal values of CNTs yield and quality mentioned in the original works.

6.4 Metaheuristic algorithms default parameters

An intensive evaluation was used for different parameter tuning settings for performance measurement purposes to find the optimal CNTs yield and quality using the best algorithms outputs. Population size and iteration number are tested among all algorithms, while some algorithms have specific parameters to be set. Table 11 to Table 14 show the different values with different settings for each parameter in each algorithm. As a result of the suggested RSM and ideal functionalisation approaches, the solubility parameter of

CNTs may be tuned, facilitating their disentanglement and distribution in a wide range of matrices. The functional groups should be strongly connected to the CNTs without disrupting the carbon substrate's structure, and as much of the CNT's surface should be left unoccupied as feasible for subsequent interactions.

6.5 Parameters setting and tuning

Some experimental tests are highly demanded to set the parameters for all algorithms to investigate the issues and behaviour changes of algorithms' performance related to parameters' tuning. Therefore, this process is implemented by testing parameters sequentially, updating the value of a single variable while holding the others constant throughout testing values. Then, the best value of the currently evaluated parameter is selected and fixed after proceeding to the next parameters testing. From the experimental result, we can elicit the effects of parameters setting on the algorithm's overall performance and the strength and weakness points of each algorithm, especially for CNTs yield and quality optimisation problems.

The eight metrics include worst, best, mean, and std. dev, mean execution time (*M_ET*), mean required iterations (*M_RI*), success rate percent (SR%), plus entail functional evaluating amount (MFEN), were compared to determine the optimal efficiency measurement for each algorithm (FEN). Thirty iterations of each tuning scenario were performed, and the average was taken. Based on the quantity and quality of replies received, the cut-off criteria used in this study were deemed to be optimum. The optimality of each method was initially determined by running a large number of iterations (up to 5,000) on a small population (100 individuals) with varying tuning without considering the running time so far (optimal value) among all algorithms.

It should be noted that each dataset has a different optimal value of CNTs output (yield/quality) due to the different solutions and objective functions. The best values or stopping conditions of the yield response in the first four cases using the tested metaheuristic algorithms in this study were found at 606.9219%, 534.93508%, 48.0118g, and 391.70259% for all case studies, respectively. While for the quality response in case study 5, the optimum minimum value was found at 0.38659. The next subsections discuss the results of sequential tuning of the algorithm parameters for all the case studies. However, the tuning process for only one case study is discussed in detail in this paper, while the results from other cases are done in the same manner as the case study 1, and the final result of the tuning process for the other cases is summarised in this paper. All algorithms' parameter settings for case studies are implemented using the objective functions declared in Table 2 with upper and lower boundaries from Table 1. All experimental tests were performed using a standard computer with a 2.13 GHz Intel Core i3 processor and 4 GB of RAM. The best setting is reported in bold font format.

6.6 Solution representation

To design a suitable solution representation for the optimisation purpose in this study, each individual solution is represented by a vector of input parameters $x_i = (x_1, x_2, \ldots, x_N)$ where N is the total number of inputs. According to this research, the overall number of input parameters for the CVD process for CNTs growing in different case studies ranges from two to four. Considering the constraints and boundary limits of each cell of the input parameter x_i in the solution, the initial solutions are generated randomly. For instance, in case study 1, there are three input variables for the CNTs growing process to represent the solution $x_i = (x_1, x_2, x_3)$, where x_i values are the factors such as catalytic mass, the vapour pressure of methanol, and heating rate, respectively, with constraints boundary limitations, i.e., $[700 \leq x_i \leq 800]$ and so on. The solution and its corresponding utility of objective value or normalised fitness value in the algorithms' memory is represented in Table 15.

$f(x_{ij})$ is calculated based on the corresponding utility of each case study.

7 Experimental results

This section discusses the tuning process and optimisation results for both carbon yield and quality in all case studies using metaheuristic algorithms.

7.1 Case study 1: chemical analysis and RSM optimisation for CNT synthesis via ammonia degradation on Co-MoAl$_2$O$_3$

The aim of this case study is to maximise the CNTs' yield. Using the reported objective function from Table 2. The stopping condition in this case study among all tested algorithms was found at a maximum yield of 606.9219%. The next subsections discuss the results of iterative enhancement of the proposed algorithms. First, however, the tuning process for only the ABC algorithm is discussed in detail; meanwhile, the tuning process for the GA, PSO, and BA algorithms was done in the same manner as ABC tuning, and the summary of all the algorithms' tuning process is reported.

7.1.1 Performance evaluation of parameters tuning in the ABC for case study 1

Tables 16 to 18 show all experimental results from the parameters tuning process for ABC. Table 16 shows the parameters tuning for the population size.

As shown in Table 16, an SR% of 100 was obtained after setting the population size to 16 and above. However, setting a population greater than 16 requires more *M_ET* and *FEN*. Therefore, among all populations which obtained 100% of SR, 16 bees are selected to be the best number in the population due to its simplicity by evaluating less FEN. Table 17 shows parameters tuning of limit.

The eight metrics include worst, best, mean, and std. dev, *MET*, *M_RI*, success rate percent (SR%), plus entail functional evaluating amount (MFEN), were compared to determine the optimal efficiency measurement for each algorithm. Table 14 shows the different values with different settings for the ABC algorithm, such as limit, population size, and iteration number. In addition, Table 17 presents the *limit* effect on the ABC performance. The best performance setting was found at *limit* = 30 with *SR*% of 100%, lowest *M_ET* of 0.025, *M_RI* of 28.067, and *FEN* of 465.1. In this case, when the solution is kept longer in the employed and onlooker bee phase before being abandoned, the performance is reduced, i.e., at *limit* = 100, while *limit* = 30 is enough to get the best performance. Finally, Table 18 shows the parameters tuning of cycles.

Table 18 shows that setting the *cycles'* number to 10 leads to trapping in local minima. The minimum required *cycle* to avoid trapping in local minima is 15. Meanwhile, 50 *cycles* could obtain 100% of *SR*%. Thus, the optimal parameter values in the ABC algorithm for this case study were found at 16, 30, and 50 for the overall population, the bound, and the convergence rate. The result from process variables for each method examined in case study 1 is tabulated in Table 19. The parameters in the column of setting values are shown sequentially.

7.2 Investigation case 2: CNT production and characterisation from polyethylene trash

The results of developments in enhanced performance across all tweaked parameters and all evaluated methods for case study 2 are summarised in Table 20. The parameters in the setting values column are shown sequentially.

7.3 Case study3: surfaces method for optimising the yield of nanocarbons prepared through chemical vapour degradation of dioxide

The overview of the findings for case study 3's performance improvement work, including tuning parameter progress and algorithm test results, is summarised in Table 21. The parameters in the setting values column are shown sequentially.

7.4 Research 4: ammonia degradation over for an alumina-based catalyst for the optimal production of CNTs

It is important to note that since the number of process parameters in case study 4 and case study 5 is three, the same as in case study 1. Therefore, the same set of the best tuning result from tested algorithms in case study 1 was generalised for case study 4 and case study 5. The results from this case study are shown in Table 22 and represent the algorithm's performance in optimising the CNTs yield.

7.5 Test case 5: ammonia oxidation over a matrix composite catalyst for the optimal synthesis of CNTs

The aim is to minimise the value ID/IG, which indicates the quality of grown CNTs. The results from all tested algorithms are summarised in Table 23. Typically, the ratio of ID/IG is used to evaluate the concentration of defects. A defect-induced double resonance scattering progression has been proposed as the origin of the G mode in CNT Raman spectra. Growing CNT arrays at low temperatures may have hidden the metallic behaviour of the CNT yarn, although this is just a superficial explanation.

8 Results analysis and validation

Referring to the tuning process, Figure 11 illustrates the algorithm's behaviours in 30 runs to improve the quantity and quality of CNTs, whereas Figure 12 depicts the behaviour of the algorithm's settings in the best possible situation studies.

In case study 1, the optimisation result showed that GA introduces a weak performance with 0% of SR% and high FEN, while other algorithms obtained the maximum target value of 607% with different SR%. ABC is the only one that efficiently could reach 100% of *SR*%; therefore the best algorithms for this case study. Figure 11(a) clearly displays that the ABC algorithm showed stability with the best performance at the peak of maximum yield value in all 30 runs with 100% of SR%; in addition, it has a low *M_ET* of 0.25 second, 28 of *M_RI*, and 465.07 of *FEN*. Since the ABC showed the best performance, the run that has the highest number of iterations at 47 iterations was selected to be illustrated in the plot in Figure 12(a) to clearly represent the constraint parameters' behaviours. Parameter values are selected based on the result from the ABC as the best-tested algorithm in this case study at population size = 16, limit = 30, and cycles = 50. In addition, obtaining optimal parameters at the earlier stage of the search process indicates that ABC performs well.

The maximum yield of ≈607 could be obtained using the ABC algorithm by setting the constraint parameters values at 761.387°C, 0.75 atm, and 0.4 g for temperature, methane partial pressure, and catalyst weight, respectively. These optimum values agree with the obtained CNTs yield in the referred original work, where the yield was optimised at 607%, meaning that the ABC algorithm, in this case, study, obtained a good result like the result from RSM in the original work.

In case study 2, PSO and GA might be trapped at local minima with insignificant performance and 0% of SR%, while the BA algorithm has achieved a satisfactory result (16.66%) in terms of SR, with enhanced quality in the exploration sections, but with low performance in exploration than GA and PSO. On the other hand, BA required 0.555 seconds and performed very high FEN at 21,379.5 times. However, it is clearly shown that ABC performs best in all eight evaluation metrics since it was the only one that obtained 100% of SR% with the best stability in mean value and std. dev. Moreover, it required a low *M_ET* at 0.071 and a low FEN of 1,970.7, related to its good exploration performance. The plot in Figure 11(b) shows the best CNTs yield obtained by 30 runs for the algorithms; GA has been omitted to enclose the values of other algorithms for clear observation. It is clearly shown that the ABC algorithm is completely stable with better performance at the peak of maximum yield value in all 30 runs.

Figure 12(b) displays the parameter values selected based on the result from the ABC as the best-tested algorithm in this case study at population size = 40 and limit = 50, and cycles = 100. ABC obtained 100% of SR%, meaning the maximum yield was obtained in all 30 runs. The behaviour of constraint parameters illustrates how changing its values improves the efficiency of the ABC in optimising CNTs yield. The maximum yield of 534.94% could be obtained by setting values of constraint parameters in the ABC algorithm at 806.66°C, 5.97 g, 150 mg, and 10 min for the factors of heat, mass, mass, and time. CNTs yield was optimised at 514% in the cited artistic creation; this research confirms a 21% increase, or a yielding of 4.07%, above that figure.

In case study 3, GA is less efficient; this could indicate that GA is trapped at local minima with 0% of SR%. PSO and BA algorithms showed better results since they require less *M_ET*, *M_RI*, and *FEN*. However, ABC has less efficiency than BA in this case study. As mentioned before in Section 3(C), this case study is very simple and has only two parameters to be optimised in its objective function variables. BA has introduced better performance among all other algorithms at 0.0048 seconds, *M_RI* of 2.37, and only 16.83 *FEN*, to conclude that BA is better in optimising simple yield optimisation cases. This could be interpreted by the shallow search space of this case study, where the algorithm does not exploit and explore much. The plot in Figure 11(c) shows the best CNTs yield obtained by each of the 30 methods in case study 3 received 30 iterations.

In this plot, PSO, BA, and ABC are stable and obtained maximum yield deposit in all 30 runs with 100% of SR, while it is clear that GA might be trapped at a local minimum fluctuating among all runs. The behaviour of the parameters in the optimal solution is illustrated in Figure 12(c); the values of the parameters are selected based on the result from the BA as the best-tested algorithm in this case study at $r_i^0 = 0.9$, $\gamma = 0.8$, $\alpha = 1$, population size = 5 and iterations = 75.

Since 30 runs have obtained the maximum yield, only one run out of them was chosen with the highest number of iterations is selected; this run number has nine iterations and selected to be clearer in the plot to represent the parameters' behaviours.

From the all obtained results in this case study, the yield has been optimised using the reported objective function with only two parameters in the referred original paper; the experimental result showed that the maximum yield of 48.01 g could be obtained by setting values of constraint parameters using BA algorithm at a temperature of 1,226.6°C and 963.3 $cm^3\ min^{-1}$ of carbon source flow rate. These optimum values are closed with the yield number in the cited primary source at which performance optimisation was performed 54.58 g. The difference between the original work and the experimental results could be interpreted by eliminating one parameter in this study: the catalyst loading, as suggested by RSM modelling work. In contrast, it was included and optimised in the referred original work, with notice that the values of the two optimised parameters using BA agree with their values in the original work using RSM.

In case study 4, it is clear that ABC has obtained the best performance with an *SR*% of 26.66%. In addition, ABC has the best performance among other algorithms at 0.039, 45.2, and 739.2 for *M_ET*, *M_RI*, and *FEN*, respectively. However, GA showed weak performance at 0% of *SR*%. PSO and BA introduced less performance than ABC at SR% of 20% for both with less efficiency than ABC. Figure 11(d) illustrates the algorithm behaviours in 30 attempts to optimise this example's original CNT study yield of CNTs. The figure also shows that BA is competitive with ABC in obtaining the optimal solution, but ABC has the best SR%, *M_ET*, *M_RI*, and *FEN* among all other algorithms. Furthermore, the run with 50 iterations was selected to clearly represent the ABC algorithm performance as in Figure 12(d).

The optimal yield of 391.70% was optimised at the maximum temperature value of 800°C, maximum time of 4hrs, and almost maximum value of metal loading at 39.44%. Better yields of CNTs were achieved with this ideal value than found in the cited original study, only 326.1%. Overall, the experimental result using ABC showed significant improvement in CNTs is effective here study compared to the original work where they used RSM. The yield has been improved here by 65.6%, which means that the improvement percentage is 20.12% of the RSM result in the original work.

In case study 5, the results show that ABC has the best performance in this case study, with the highest SR% of 73.33% and a low *M_ET* of 0.027 seconds. In addition, it has the best *M_RI* and *FEN*. Moreover, Figure 11(e) shows the algorithm's behaviours in 30 runs to maximise the quality by minimising the ID/IG ratio; GA showed weak performance, while other algorithms presented better results. ABC obtained the best SR% of 73%, while BA fluctuated at a few points with an SR% of only 20% around the optimal solution. The run with 49 iterations was selected to represent the algorithm performance clearly on the plot in Figure 12(e).

The experimental tests using ABC found the optimal ID/IG value. It can be realised that the optimal value of 0.387 is predicted to reach its heat input of 800°C, the maximum duration of 4 hours, and the minimum value of metal loading of 10.185%. CNTs with this optimal value were of greater quality than those in the cited study, in which ID/IG was tuned at 0.609%.

The finding from this case study reveals that the experimental result using ABC could help to find better ID/IG values than using RSM. Furthermore, the CNTs quality has been improved by minimising the ID/IG value with 0.222, which means that the quality improved by 36.45% better than the original work where RSM was used.

The values of the ideal growth conditions for optimum quality and yield are translated into the optimal solutions or multiple regressions listed in Table 2 to verify the optimal outcome from the algorithms in this research. The model's predicted outcome and the optimised solution should be identical. Inputting the estimated optimum growth conditions from the suggested algorithms into a regression model yielded the same anticipated CNTs yield and quality. This suggests that similar findings might be achieved in a real experiment scenario utilising CVD to produce CNTs. The presented algorithms in this work may thus be efficient optimisation techniques in future investigations with comparable aims.

In conclusion, the algorithms' performance is ordered as *ABC* > PSO > BA > GA, *ABC* > BA > PSO > GA, *BA* > PSO > ABC > GA, *ABC* > PSO > BA > GA, and *ABC* > PSO > BA > GA for experiment 1, experiment 2, experiment 3, experiment 4, and experiment 5, respectively. ABC showed that it is the most appropriate selected metaheuristic algorithm to optimise CNTs yield and quality. This finding could help researchers to use the ABC algorithm in similar industrial problems. Moreover, the tuning process performed in the earlier stage of this study had a great impact and clearly enhanced the outputs of the algorithms. Therefore, this process with its settings is also highly recommended to evaluate the performance of a new proposed metaheuristic algorithm.

CNT synthesis is emerging with technological advancement due to its broader applications. Currently, CNTs are being synthesised by chemical as well as physical approaches. This research has shifted towards developing CNTs-based nanomaterials using the RSM model due to their advanced and unique chemical, electronic, mechanical, and structural features. Nanocomposite has become the backbone of all scientific industries due to their advanced mechanical, electrical, thermal, optical, catalytic, and electrochemical properties, making them remarkably dissimilar from component materials. Considering the progress in laser ablation and electrical arc discharge methods, significant advancement has been made in the large-scale production of CNTs.

9 Optimisation's impact on the quality of the final product of Synthe of CNTs

For CNTs yield optimisation, it was found that metal loading, catalyst weight, the relative concentration of gas, polymeric density, and carbon source flow rate are effective at high levels. On the other hand, for the case of CNTs, quality, reaction temperature, and duration were found better at a high level, while metal loading was found better at a low level.

To investigate the optimisation result, the optimised constraint parameters in this paper are analysed and supported by previously published works as follows:

- In case study 1, the upper boundaries of the partial pressure and catalyst weight were obtained, while the optimal temperature was obtained at 761°C. The empirical result in the original work showed that the yield decreased between 760°C and 800°C due to severe catalyst deactivation; this result agrees with another previously reported result by Chen et al. (2004). Moreover, at the optimum process parameters settings, it was observed that the carbon dioxide concentration rises with increasing gas pressure head. Diffusion precipitation on catalytic and then increases the yield of CNTs proportionality and resulted in the formation of polymeric carbon species, while a catalyst weight effect was found moderate on CNTs yield (Chai et al., 2011). On the other hand, by setting the CNTs synthesis process at these optimal parameters with 8Co – $2Mo/Al_2O_3$ catalyst, the grown CNTs have a nearly uniform diameter with 11.8 nm to confirm growing MWCNTs with open-tip morphology. However, it is worth mentioning that defects and amorphous carbon were found in the structure of the CNTs wall at these optimum values (Chai et al., 2011).
- In case study 2, the ABC result showed that at a polymer weight of 5.97 g with 806.66°C and 150 mg of catalyst weight for 10 min, the CNTs yield is expected to be increased. However, a higher polymer weight of more than 5 g at 800°C and less catalyst weight of 100 mg do not increase CNTs yield (Bajad et al., 2015). On the other hand, constraint parameters resulting from ABC are supported by other studies; for example, Li et al. (2005) demonstrated that at a higher amount of catalysts weight at 150 mg, a dissociation of hydrocarbons is more efficient to an individual carbon atom. Also, the temperature at 806.66°C is appropriate to increase CNTs yield and in agreement with the result in the original paper where they mentioned that at higher temperatures of 850°C and 900°C, catalyst activity is decreased, and the hydrocarbon source for CNT production is minimised due to rapidly passing out the hydrocarbon vapours (Chen et al., 2004). Moreover, at a short time of 10 min, the CNTs increase as proof that an increase in time decreases carbon yield (Bajad et al., 2015).
- In case study 3, the optimum result from BA found that the best constraints parameters setting was detected at elevated values for all temperature and carbon source flow rate process parameters. This result agrees with the original work by (Allaedini et al., 2016), where they implemented RSM and mentioned that elevated temperatures significantly affect CNTs yield. Furthermore, it was found that the temperature above 1,100°C only applies to growing CNTs. However, the amorphous CNTs, i.e., cubes and chunks, are grown at lower temperatures at 700°C and 900°C (Zhang et al., 2013), which requires a purification process, meaning that simple control of CNT's reaction temperature leads to the synthesis of different morphologies of CNTs. In addition, at an elevated temperature, a diameter of 8.45 nm was observed to confirm the formation of MWCNTs with a bamboo-herringbone structure (Allaedini et al., 2016) The diameter of MWCNTs is ranged between 2 to 30 nm, while 0.4 to 2 nm for SWCNTs (Eatemadi et al., 2014). However, a few trapped catalyst powders are observed on the grown CNTs after purification at a high temperature of 1,100°C (Allaedini et al., 2016).
- The analysis of case study 4 and case study 5 was already discussed and supported with related work in Section 5.4.

10 Conclusions

This study discussed the modelling and optimisation of the CNTs' yield and quality. The CNTs in the five selected case studies were grown using the CVD method. RSM was employed in the experimental design and modelling work for all the case studies. Besides developing regression models using RSM, the data and models were statistically analysed, and the significant CVD process parameters for the CNTs growth were identified. Further, its effect on the CNT's yield and quality was discussed. The regression models from RSM were used as the objective functions for optimisation using four metaheuristic algorithms, i.e., GA, PSO, BA, and ABC. The optimisation process used efficient parameters tuning strategy to improve the algorithm's performance and output. The findings from the modelling and optimisation in this study were also discussed and supported by other researchers from the real-world applications of the CNTs growing using the CVD process. The mechanical properties of the macroscopically long and continuous CNT have been directly measured for the first time in this study. Based on our research and the available literature, this is the first report of topological indices for this specific type of CNT interconnections using the RSM model and is widely applicable in the academic world.

The findings from this paper concluded that RSM is an efficient way to model and analyse CNTs' yield and quality responses. In addition, the tested metaheuristic algorithms were very efficient in finding the optimal CNTs yield and quality in all case studies. Among the tested algorithms, the results concluded that ABC performs best in most of the case studies. Furthermore, BA could be used to find the optimal CNTs yield in the simple case study presented in case study1 with only two input process parameters. Therefore, the proposed metaheuristic algorithms could improve the result for better CNTs yield and quality and found better results than RSM result in the original referred works. Besides, ABC could be generalised for CNTs yield and quality optimisation problems and used in other manufacturing processes. Also, the findings from this result reveal that using metaheuristic algorithms can address the main issues of the CNTs synthesis and manufacturing process, which are the cost and customisation toward green and sustainable manufacturing. Optimising the carrier gas, flow rate, catalyst/carbon-source ratio, and catalyst type for aligned CNT synthesis utilising a two-stage catalytic CVD setup is possible and may be reported in the future.

Tables and Figures are available on request by emailing the corresponding author.

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