Using Box-Behnken Design for Determination of Antimony(III) in Polyethylene Terephthalate by Inductively Coupled Plasma-Optical Emission Spectroscopy

Nur Faten Hazira Mohd Ghazali[1], Nor Suhaila Mohamad Hanapi[1*], Nurzaimah Zaini[1], Ahmad Lutfi Anis[2], and Wan Mohd Asyraf Wan Mahmood[3]

[1]*Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia*

[2]*Faculty of Applied Sciences, Universiti Teknologi MARA, 94300 Kota Samarahan, Sarawak, Malaysia*

[3]*Center of Foundation Studies, Universiti Teknologi MARA, 43800 Dengkil, Selangor, Malaysia*

Corresponding author:

tel: +603-55435744
email: norsuhaila979@uitm.edu.my

Received: March 29, 2025
Accepted: June 21, 2025

DOI: 10.22146/ijc.105759

Abstract: *Antimony (Sb), a toxic heavy metal, is widely used in plastic industries as a catalyst in producing polyethylene terephthalate (PET) due to its high catalytic efficiency and cost-effectiveness. However, its potential to leach into consumables raises significant health and environmental concerns, highlighting the need for accurate quantification methods. This study aimed to optimize microwave-assisted digestion (MW-AD) parameters for the determination of trace levels of Sb(III) in PET using inductively coupled plasma-optical emission spectroscopy (ICP-OES). A Box-Behnken Design (BBD) was employed to systematically optimize three critical factors: digestion temperature, digestion time, and sample weight. The statistical analysis confirmed the model's reliability, with an R^2 value of 0.9945, indicating a well-fitted model. The optimal conditions were determined as 200 °C digestion temperature, 15 min digestion time, and 0.1 g sample weight. Under optimal conditions, the method exhibited excellent analytical performance, achieving a limit of detection (LOD) of 1.079 mg L^{-1}, a limit of quantification (LOQ) of 3.270 mg L^{-1}, accuracy of 106% spike recovery, and precision with 0.81% relative standard deviation (RSD, n = 3). This validated method provides a rapid, sensitive, and efficient approach for the quantification of Sb(III) in PET samples.*

Keywords: *microwave-assisted digestion; antimony(III); Box-Behnken; PET; ICP-OES*

■ INTRODUCTION

Plastics play a crucial role in the present society, serving as essential materials for various consumer products, including food and beverages, and healthcare products. Among the different types of plastics, polyethylene terephthalate (PET) has gained prominence due to its excellent mechanical strength, thermal stability, and cost-effectiveness [1]. PET is widely used in the packaging of food and beverages, particularly in bottled water and soft drinks, due to its transparency, lightweight nature, and recyclability [2]. However, concerns have been raised regarding the presence of heavy metals in PET, particularly antimony (Sb), which is commonly used as a catalyst in PET production [3].

Heavy metals in plastics are not chemically bound to the polymer matrix but instead embedded within it, making them susceptible to leaching under environmental conditions such as heat, pH variations, and prolonged storage [4]. Antimony poses potential health risks as it can migrate from PET containers into food and beverages, especially under elevated temperatures [5]. The European Commission (EC) has established regulatory limits for antimony migration in food, setting a maximum allowable concentration of 40 μg kg^{-1} in food containers and 5 μg L^{-1} in drinking water (EC Directive 2020/1245) [6]. Consequently, the accurate quantification of antimony in PET is essential to ensure consumer safety and regulatory compliance. Studies have shown that various factors, including storage duration, temperature fluctuations, and beverage composition, can influence the leaching of antimony into bottled liquids [7].

Various digestion methods have been employed for the quantification of heavy metals in polymer-based samples, including food-contact materials such as PET. Conventional approaches such as wet acid digestion, dry ashing, and alkaline digestion have been widely used; however, each presents specific limitations [8-10]. Wet acid digestion, while simple, is time-consuming and prone to contamination due to its open-vessel nature. Dry ashing is effective in removing organic matter but may cause analyte loss through volatilization, whereas alkaline digestion often results in incomplete sample breakdown and is only suitable for certain polymer types. Microwave-assisted digestion (MW-AD) is considered more efficient in terms of processing time, solvent usage, and temperature control, and is viewed as a more sustainable technique [11]. In addition, it provides enhanced analyte recovery, minimizes reagent consumption, and ensures safer operation using closed vessels [12-13].

Despite these advantages, precise optimization of MW-AD parameters such as digestion temperature, time, and sample weight are essential to prevent overpressure, ensure complete digestion, and achieve high analytical precision [14]. However, the lack of a standardized optimization approach in current studies has led to inconsistent results. Therefore, a systematic and well-optimized digestion strategy is critical for improving the reliability and reproducibility of trace metal analysis in polymer matrices.

This study aims to optimize microwave-assisted digestion parameters for the determination of Sb(III) in PET using ICP-OES. This research may contribute to the development of a rapid, efficient, and reliable method for the quantification of antimony in PET matrices. Optimizing MW-AD conditions may lead to minimized sample preparation time, reduced reagent consumption, and enhanced analytical sensitivity. The findings of this study may assist in the establishment of improved analytical protocols to ensure consumer safety and compliance with environmental and health standards. Furthermore, this methodology can be extended to the analysis of other heavy metals in plastic packaging materials, supporting broader environmental monitoring efforts.

■ EXPERIMENTAL SECTION

Materials

Analytical grade 65% nitric acid and 35% hydrogen peroxide (Fisher Scientific, Malaysia) were used for sample digestion. Standard Sb(III) solutions (1,000 mg L^{-1}) were obtained from Perkin–Elmer for ICP-OES analysis. All dilutions and sample preparations were performed using Milli-Q™ double-distilled water (conductivity < 0.055 µS/cm).

Instrumentation

Sb(III) determination was performed using an ICP-OES (Perkin–Elmer Optima 2100 DV) equipped with a dual-view plasma torch for axial and radial detection. The sample introduction was achieved via a pneumatic nebulizer and an AS-70 autosampler for automated handling. The system was optimized for Sb(III) detection at 217.581 nm [15], with key operating parameters as listed in Table 1.

Microwave-assisted digestion (MW-AD) was conducted using a Milestone Ethos Easy (MA174-001) oven equipped with a temperature and pressure control system. The Milestone Ethos Easy (MA174-001) oven enables precise regulation of the digestion environment, ensuring efficient and safe sample processing [16]. The system has the ability to monitor and adjust pressure atomically provides a critical safety feature, preventing vessel rupture and ensuring consistent digestion conditions across multiple samples [17]. In this study, six PTFE digestion vessels were applied in the system or sample processing. The use of PTFE digestion vessels is crucial due to their chemical inertness and ability to withstand the elevated temperatures and pressures generated during microwave digestion [18].

Table 1. ICP-OES Operating parameters for Sb(III) determination

Parameter	Variable	Data (L/min)	Data
Wavelength	217.581 nm	Plasma gas flow	15.00
RF power	1150 W	Auxiliary gas flow	0.80
Pump speed	50 r/min	Nebulizer gas flow	0.75
Back pressure	117.0 kPa	Sample uptake rate	1.50

Procedure

Preparation of standard and sample collection

A PET food container was procured from the local Eco store. The samples were cleaned with tap water and domestic detergent, followed by rinsing with deionized water to ensure complete cleaning and removal of paper or plastic labels. The samples were then sectioned into smaller pieces (less than 1 cm^2) and air dried. To ensure the homogeneity and complete digestion of the sample before analysis, samples were then blended to a smaller size using a home blender.

MW-AD procedure

The procedure for MW-AD adopts the method established by Goodlaxson and co-workers [19] to determine Sb(III) in PET materials. The schematic for the MW-AD procedure is shown in Fig. 1. In this method, a weighed PET plastic sample is placed in a PTFE tube (approximately 15–20 cm long and 3–5 cm in diameter) before adding 6 mL of HNO_3 and 2 mL of H_2O_2. The sample is gently swirled to mix it with the acids before securing the inner cap and locking the outer vessel jacket. The vessels are then placed on the microwave rotor with a sensor inserted into a blank vessel to monitor the digestion conditions. The digestion process involves three steps. In the first step, the system is set to 90 °C for 2 min at 800 W. The second step increases the temperature to 180 °C for 10 min while maintaining the same power level. The final step is determined based on the experimental design model, using a power of 800 W. Subsequently, the solutions were cooled, filtered to remove any undissolved residue, and diluted to a final volume of 15 mL using deionized water. The Sb(III) concentration in the filtrate is then measured using standard solutions ranging from 0.0 to 10.0 mg L^{-1}.

Experimental design

MW-AD-ICP-OES conditions were optimized using a Box Behnken Design (BBD) to obtain the optimum conditions for the simultaneous extraction of Sb(III). RSM and BBD were used to optimize the three independent variables (sample weight, temperature, and extraction time). Finally, the experimental design was generated using the software for regression analysis of the experimental data to fit the equations.

Statistical data and analysis

An Analysis of Variance (ANOVA) was applied to the experimental data and the results. These statistical analyses and BBD were performed using Design Expert State-Ease software, version 13. The experimental variables to be tested were randomly designed by BBD.

Validation of analytical method

The validation of the MW-AD analytical method was conducted to ensure its reliability and suitability for the intended purpose. The parameters assessed included linearity (R^2), precision, accuracy, limit of detection (LOD), and limit of quantification (LOQ). The LOD and LOQ were determined using the standard error of the intercept from the calibration curve. The LOD was calculated by multiplying the standard error of the intercept by 3.3, while the LOQ was obtained by multiplying the standard error of the intercept by 10, both divided by the slope of the calibration curve. The precision of the method was expressed in terms of relative standard deviation (RSD%) and accuracy in terms of spike recovery. Such equations are referred to as Eq. (1).

$$\text{Recovery}(\%) = \left(\frac{\text{Unspike} - \text{Spike}}{\text{Known concentration added}} \right) \times 100\% \quad (1)$$

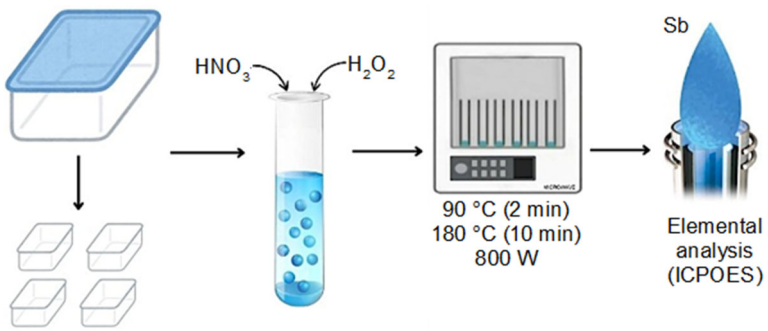

Fig 1. Schematic of the microwave-assisted digestion (MW-AD) procedure

■ RESULTS AND DISCUSSION

Optimization of MW-AD Parameters Using BBD

RSM is a practical quantitative research technique that uses mathematical and statistical software to explore the relationships and interactions between multiple input parameters and responses [20]. Three parameters, namely sample weight (A), microwave heating temperature (B), and microwave heating time (C), were optimized and analyzed using BBD. A total of 15 runs were conducted, and the results are summarized in Table 2.

Statistical Analysis, Model Fitting, and Regression Analysis

The effectiveness of BBD model in optimizing MW-AD parameters was assessed using ANOVA. This statistical analysis evaluates the significance of the model by examining p-values, sum of squares, mean square, F-values, lack-of-fit test, and degrees of freedom. The ANOVA results are summarized in Table 3. The model demonstrated a strong predictive capability with a statistically significant high F-value of 60.67 and a very low probability (0.31%) of it occurring due to random noise. In addition, the Lack of Fit has an F-value of 4.57 and was found to be statistically insignificant ($p = 0.166$), indicating that the model fits experimental data correctly, as it implies that the unexplained variation is random rather than systematic errors in the model.

The result confirms the significant impact of the independent variables, that is, sample weight (A), temperature (B), and time (C), on the digestion efficiency of antimony in PET. This significance is proven by the low p-values (< 0.05) and high F-values associated with each factor [21]. Among them, time (C) stands out as the most individually significant, with the highest F-value (304.40), followed by temperature (B) with an F-value of 246.35. These findings highlight the importance of optimizing time and temperature to maximize digestion efficiency. In addition, the statistical analysis of the model highlighted both linear and non-linear effects of the factors and their interactions.

According to the p-values, temperature (B^2) and sample weight (A^2) were identified to have significant negative quadratic terms, indicating non-linear relationships. This suggests the presence of optimal conditions for temperature and sample weight, where any excessive increase or decrease beyond specific points may negatively impact the digestion efficiency. In contrast, the quadratic term for time (C^2) was small and insignificant (0.1149), suggesting a straightforward linear effect of time on the response within the tested range.

Table 2. The Box-Behnken design runs for Sb by MW-AD-ICP-OES

Run	Factors			Response
	A: Sample weight (g)	B: Temperature (°C)	C: Time (min)	Sb concentration (mg L^{-1})
1	0.300	210	15	2.590
2	0.300	200	20	2.782
3	0.100	210	20	0.955
4	0.500	200	15	3.200
5	0.300	200	20	3.011
6	0.500	210	20	3.555
7	0.500	190	20	3.066
8	0.100	190	20	1.060
9	0.300	200	20	2.872
10	0.500	200	25	1.132
11	0.100	200	25	0.865
12	0.300	210	25	3.541
13	0.300	190	25	3.143
14	0.300	190	15	2.777
15	0.100	200	15	4.436

Table 3. ANOVA analysis

Source	Sum of squares	DF	Mean square	F-value	*p*-value	Significant
Model	15.9100	9	1.7700	60.6700	0.0031	***
A-Sample weight	0.4109	1	0.4109	14.1000	0.0330	*
B-Temperature	7.1800	1	7.1800	246.3500	0.0006	***
C-Time	8.8700	1	8.8700	304.4000	0.0004	***
AB	4.6300	1	4.6300	158.7800	0.0011	***
AC	0.5648	1	0.5648	19.3800	0.0217	*
BC	5.4600	1	5.4600	187.3500	0.0008	***
A^2	0.9995	1	0.9995	34.3000	0.0099	**
B^2	5.0900	1	5.0900	174.7900	0.0009	***
C^2	0.0373	1	0.0373	1.2800	0.3404	-
Residual	0.0874	3	0.0291			
Lack of Fit	0.0608	1	0.0608	4.5700	0.1660	-
Pure Error	0.0266	2	0.0133			
Cor. Total	16.0000	12				

Note: "-" means no significance; "*" means significant ($p < 0.05$); "**" means moderately significant ($p < 0.01$), and "***" means extremely significant ($p < 0.005$)

Furthermore, the interaction effects, that is, sample weight and temperature (AB), temperature and time (BC), and sample weight and time (AC), were also found to be statistically significant with *p*-values less than 0.05. These results indicate that the combined effects of these factors play a crucial role in determining digestion efficiency, emphasizing the interdependence between factors.

The final regression equation of the fitted model is shown by Eq. (2), where Y is the response (concentration) of Sb(III) analytes, A is sample weight, B is digestion temperature, and C is heating time.

$$Y = 2.89 - 0.2926A + 1.64B - 1.3C + 1.64AB + 0.3758AC + 1.78BC - 0.5950A^2 - 1.63B^2 + 0.1149C^2 \quad (2)$$

The regression analysis demonstrates how well the quadratic model fits the response data [22]. The model shows a high coefficient of determination (R^2) of 0.9945, indicating that it explains 99.45% of the variability in the response. The adjusted R^2 value of 0.9811 further confirms the model's ability to explain the variance while accounting for the number of terms in the model. Although the predicted R^2 is unavailable due to high-leverage data points (reported as 1.0000), the model's reliability is supported by an adequate precision value of 23.25, which far exceeds the minimum acceptable threshold of 4. The coefficient of variation (C.V.) of 6.47% reflects the model's precision and reproducibility in describing the response. Based on this validated quadratic model, the optimal conditions for MW-AD of Sb(III) from PET were identified: a sample weight of 0.1 g, a digestion temperature of 200 °C, and a heating time of 15 min.

The ANOVA and regression analyses were conducted under key assumptions, including normality of residuals, independence of observations, and homogeneity of variance. Diagnostic checks using residual and normal probability plots (not shown) indicated that these assumptions were adequately met. Furthermore, the low pure error and non-significant lack-of-fit confirm the adequacy of the model.

It is important to note some limitations of the BBD employed in this study. While BBD is efficient and reduces the number of experimental runs, it does not include extreme factor combinations (corners of the design space), which may limit the ability to detect certain nonlinear or interaction effects at these boundaries. Additionally, the limited number of center points may affect the robustness of error estimation. Despite these limitations, the high R^2, adjusted R^2, and adequate precision values support the model's reliability and predictive power within the experimental domain tested.

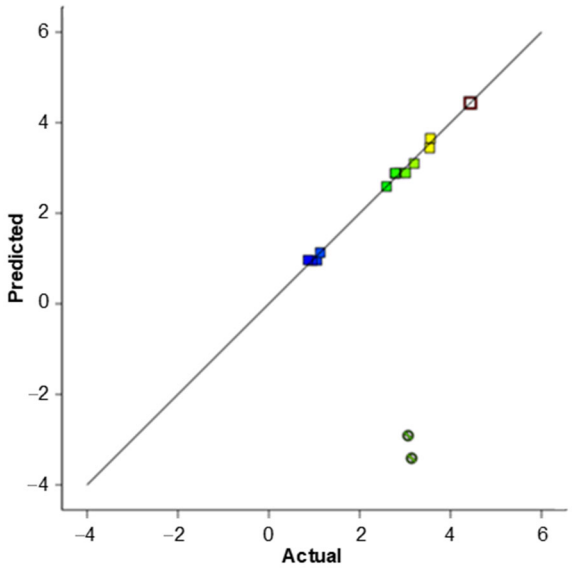

Fig 2. Predicted versus actual concentrations of antimony

Predicted and Expected Values

Comparison between predicted and actual concentrations is crucial to evaluate the accuracy and reliability of the developed model [23]. Fig. 2 illustrates the predicted versus actual concentrations of Sb(III) obtained during the optimization process. The diagonal line represents the ideal scenario where predicted and actual values are identical. Most data points were closely aligned with this line, indicating the model's strong predictive accuracy within the experimental range.

Interaction Effects and 3D Surface Plot Analysis

The interaction effects between parameters play a critical role in determining digestion efficiency in MAD. Unlike individual factors, which operate independently, interactions such as sample weight and temperature (AB), temperature and time (BC), and sample weight and time (AC) reveal how the combined influence of two factors can significantly alter digestion outcomes, as evidenced by their strong statistical significance (F = 158.78, 19.38, and 187.35; p = 0.0011, 0.0217, and 0.0008, respectively). The significant interaction effects suggest that the influence of one factor on digestion efficiency depends on the level of another [24]. This shows the importance of evaluating interactions and individual factor contributions together to achieve a more comprehensive understanding of digestion. Fig. 3 visualizes the effect of two interacting factors on the Sb(III) concentration, with the third factor held constant. The plots show the interactions between (a) sample weight and temperature, (b) sample weight and time, and (c) temperature and time. The response (Sb concentration) is represented on the z-axis, with the x- and y-axes showing the interacting factors.

Fig. 3(a) shows the effect of sample weight and temperature (AB) on the Sb concentration extracted during microwave-assisted digestion. The response surface shows that Sb(III) concentration increases with rising temperature [25]. The steep gradient along the temperature axis suggests that temperature has a more dominant effect than sample weight. However, excessive sample weights, such as 0.5 g, slightly reduced efficiency, likely due to reagent saturation or incomplete digestion. The interaction graph supports this observation, showing that at lower sample weights (0.1 g), the Sb(III) concentration increases sharply with rising temperature. The increase is less pronounced at higher sample weights, suggesting diminished temperature effects due to slower digestion kinetics or reagent limitations. The model predicts optimal conditions are 0.1 g sample weight and 200 °C temperature, where maximum digestion efficiency is achieved. These findings align with the contour plot, highlighting the need to balance sample weight and temperature for effective digestion without overloading the system.

Fig. 3(b) illustrates the interaction between sample weight and digestion time (AC) on the extraction of Sb(III) during microwave-assisted digestion. The contour plot shows that Sb(III) concentration increases with longer digestion times, especially with higher sample weights, showing the need for an extended digestion period to process the matrix fully [26]. In contrast, the response is relatively constant over the digestion time range for lower sample weights (0.1 g). This suggests minimal dependence on time due to more straightforward digestion, meaning that shorter time (15 min) are sufficient for efficient digestion. Thus, adjusting digestion time based on sample size is essential for consistent and reliable results.

Fig. 3(c) shows the interaction between temperature and digestion time (BC) on the recovery of Sb(III) during

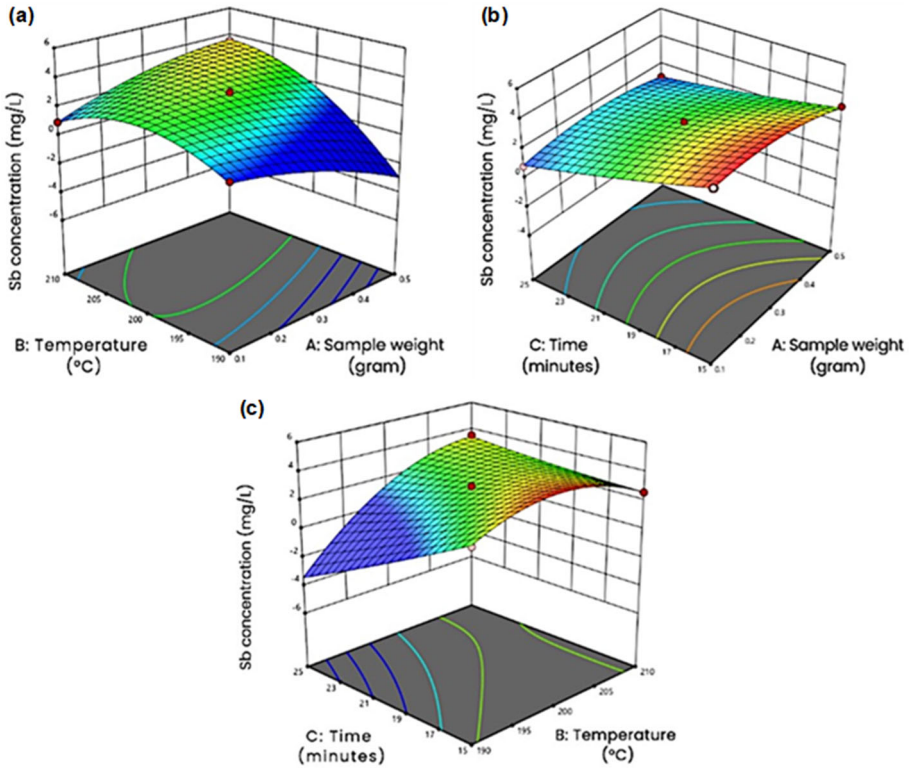

Fig 3. 3D surface plots illustrating the interaction effects of (a) sample weight and temperature (AB), (b) sample weight and time (AC), and (c) temperature and time (BC) on Sb concentration during microwave-assisted digestion

MAD. The response surface shows a curved contour, where Sb(III) recovery increases significantly as temperature and time approach their optimal levels. The highest recovery is observed at moderate temperatures (200 °C) and minimal digestion times (approximately 15 min). However, the concentration decreases in extreme conditions, such as very high temperatures and long digestion times. This might be due to Sb(III) being lost through evaporation or breaking down due to over-digestion [27]. Statistical analysis confirms this interaction as the most significant among the other interaction factors, as reflected by its high F-values and low *p*-values. This highlights the importance of carefully controlling temperature and time as they work together to improve Sb(III) recovery more effectively than when considered separately.

Pareto Chart

The Pareto chart (Fig. 4) shows which factors and interactions have the biggest impact on Sb(III) recovery during microwave-assisted digestion based on their F-values. This statistical measure revealed which variables are the most critical for optimization [28]. Digestion time (C) has the highest F-value, meaning it has the most influence, followed by temperature (B) and their interaction (BC). These results highlight that both time and temperature are the keys to achieving good digestion. This is because digestion time is crucial to ensuring the complete breakdown of the sample matrix, while temperature plays a key role in accelerating digestion. In contrast, sample weight (A), its quadratic term (A^2), the interaction between sample weight and digestion time (AC), and the quadratic effect of digestion time (C^2) have little effect.

Analytical Performance

The LOD and LOQ were calculated using the formula based on the standard error (SE) of the intercept and the slope from the calibration curve [29]. In addition, this method can reflect the variability or uncertainty of the regression model. Based on the linear regression method calculation of LOD and LOQ, the values obtained were 1.079 and 3.270 mg L^{-1}, respectively.

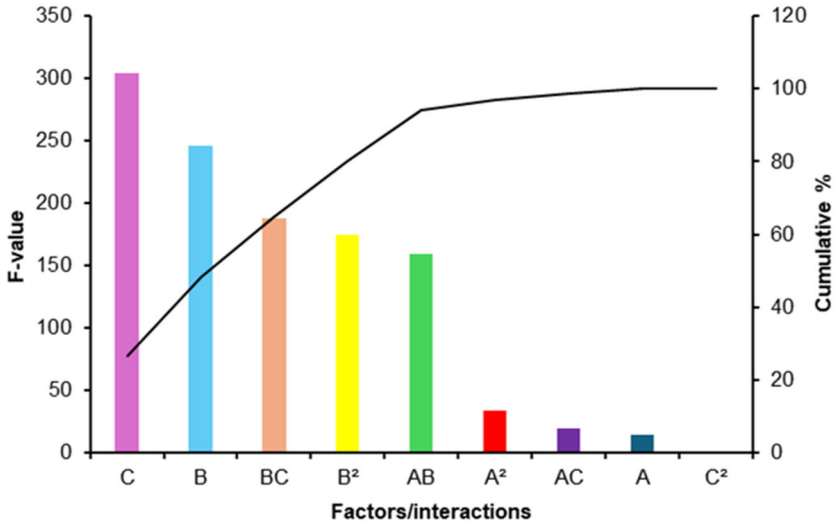

Fig 4. Pareto chart on Sb(III) recovery

The LOD value represents the smallest concentration of antimony that can be reliably detected but not necessarily quantified. In this study, an LOD of 1.079 mg L^{-1} shows that the method is sensitive enough to detect very small amounts of antimony in the sample. On the other hand, the LOQ represents the lowest Sb(III) concentration that can be accurately quantified. LOQ of 3.270 mg L^{-1} indicates that quantifiable levels of Sb(III) can be measured reliably starting from this concentration. The relative standard deviation (RSD) for triplicate determination of the unspiked Sb(III) sample was obtained as 0.81%. Table 4 demonstrates the overall analytical performance of the optimized method.

The calculated spike recovery was 106%, which falls within the generally acceptable range of 80–120%, confirming the method's accuracy. The high percentage recovery shows that digestion effectively minimizes matrix interference and analyte loss, ensuring reliable trace analysis. These findings validate the method's robustness and suitability for quantifying antimony in PET, proving its reliability for trace element analysis in complex matrices.

Comparison of MW-AD-ICP-OES with Other Reported Methods of Sb Quantification

The microwave-assisted digestion method proposed in this study was compared with other published methods for antimony analysis in PET, as shown in Table 5. Various digestion techniques, such as alkaline methanolysis, conventional wet acid digestion,

Table 4. Analytical performance of the optimized method

Analytes	Linearity (mg L^{-1})	Correlation of coefficient (R^2)	%RSD (n = 3)	Spike recovery (%)	LOD (mg L^{-1})	LOQ (mg L^{-1})
Antimony	0.0–10.0	0.9885	0.810	106.000	1.079	3.270

Table 5. Comparison of analytical techniques for Sb quantification in PET samples with other studies

Sample	Method	Analyte	Detection method	LOD (µg L^{-1})	Recovery (%)	Ref.
PET plastics	Microwave-assisted digestion	Sb(III) Sb(V)	FC-ICP-MS	0.0005 0.0007	93.0 ± 9 95.0 ± 5	[32]
PET bottles	Alkaline methanolysis	Sb(III)	HG-MP-AES	3000.0000	93.8%–99.3	[34]
PET water bottles	Conventional wet acid digestion	Sb	GFAAS	0.5000	90.0–110.0	[30]
PET	HPA	Sb	ICP-OES	60.0000	96.1–101.4	[31]
PET containers	Microwave-assisted digestion	Sb	ICP-OES	1079.0000	106.0	This study

high-pressure ashing (HPA), and microwave-assisted digestion, have been coupled with various analytical techniques, such as ICP-OES, inductively coupled plasma mass spectroscopy (ICP-MS), hydride generation microwave plasma atomic emission spectroscopy (HG-MP-AES), and graphite furnace atomic absorption spectroscopy (GFAAS). Each approach has its own advantages and limitations in terms of sensitivity, accuracy, efficiency, and practicality.

In terms of efficiency and practicality, the MW-AD-ICP-OES method offers fast sample preparation (approximately 30 min total digestion time), consumes smaller volumes of acid, and ensures safer, closed-vessel digestion. This minimizes both reagent usage and hazardous waste generation. It also avoids the prolonged digestion times and manual handling required by conventional wet acid digestion or HPA methods. The system relies on widely available equipment and reagents, making it suitable for routine use.

From a cost perspective, ICP-OES is more accessible and affordable than ICP-MS, which, although highly sensitive with detection limits as low as 0.0006 μg L^{-1} [30] requires significant capital investment, higher operational costs, and specialized laboratory infrastructure. In contrast, the ICP-OES technique used in this study, with a detection limit of 1079 μg L^{-1}, provides adequate sensitivity for regulatory monitoring at a much lower cost. Furthermore, it allows multi-element detection and does not require element-specific light sources, unlike techniques such as GFAAS.

In terms of accuracy, the developed method demonstrated a spike recovery of 106%, well within the generally acceptable range of 80–120%. This compares favorably with conventional wet acid digestion (90–110%) and other microwave-assisted methods (96.1–101.4%) [31-32]. These results confirm the method's robustness and reliability for the determination of antimony in PET matrices.

Regarding instrumental limitations, GFAAS offers lower detection limits (~0.5 μg L^{-1}) than ICP-OES but depends on hollow cathode lamps (HCLs), which are element-specific and not always easily available. This limits its flexibility and increases operational complexity. In contrast, ICP-based techniques like ICP-OES and ICP-MS are more versatile and efficient for multi-element analysis, without the need for specialized lamps [33]. Meanwhile, HG-MP-AES, although applied in PET antimony analysis, has a relatively high detection limit (3000 μg L^{-1}), making it less suitable for trace-level quantification [34].

The findings of this study demonstrate that the antimony concentrations measured in PET samples fall well below established safety thresholds. According to the European Community (EC), the directive limit is 5 μg L^{-1} for antimony in drinking water (Directive 98/83/EC) and 40 μg kg^{-1} for PET-packaged foods (Regulation (EU) No 10/2011). Beyond these, global standards such as the World Health Organization's guideline of 20 μg L^{-1} for drinking water, as well as limits set by Health Canada and regulatory agencies in Australia and New Zealand, were also considered [35-37]. The optimized method consistently produced Sb concentrations within these acceptable limits, confirming its suitability for both regional and international regulatory compliance. Although this method is less sensitive than ICP-MS, it offers a strong balance between sensitivity, accuracy, cost-effectiveness, and ease of use, making it well-suited for routine monitoring of antimony in PET containers. Future incorporation of ICP-MS may enhance detection at trace levels, but the present method already represents a practical and reliable tool for safety assessments related to PET materials.

■ CONCLUSION

MW-AD parameters for the determination of Sb(III) in PET using BBD for the optimization and ICP-OES were successfully optimized. The optimal conditions were determined to be a digestion temperature of 200 °C, a digestion time of 15 min, and a sample weight of 0.1 g. Under these conditions, the method demonstrated excellent analytical performance, low sensitivity, good accuracy, and precision, confirming its reliability for antimony quantification in PET matrices. These findings may contribute to improving food safety and environmental monitoring, addressing concerns related to antimony leaching from PET containers. The

results of this study are particularly significant as they demonstrate compliance with current regulatory guidelines, reinforcing the reliability and accuracy of the optimized method and providing a robust framework for effective monitoring of Sb(III) levels in PET materials.

■ ACKNOWLEDGMENTS

The authors would like to thank Universiti Teknologi MARA (UiTM), Shah Alam, Selangor, for providing research facilities and the Ministry of Education Malaysia for financial support through the FRGS research grant 600-IRMI/FRGS 5/3 (010/2019).

■ CONFLICT OF INTEREST

The authors have no conflict of interest.

■ AUTHOR CONTRIBUTIONS

Nur Faten Hazira Mohd Ghazali conducted the experiment, Nor Suhaila Mohamad Hanapi analyzed, revised the manuscript and was responsible for submission, Nurzaimah Zaini analyzed the raw data, Ahmad Lutfi Anis revised the manuscript and Wan Mohd Asyraf Wan Mahmood analyzed the validation of extraction data. All authors agreed to the final version of this manuscript.

■ REFERENCES

[1] Wang, K., Guo, C., Li, J., Wang, K., Cao, X., Liang, S., and Wang, J., 2024, High value-added conversion and functional recycling of waste polyethylene terephthalate (PET) plastics: A comprehensive review, *J. Environ. Chem. Eng.*, 12 (5), 113539.

[2] Chowdhury, T.U., Mahi, M.A., Haque, K.A., and Rahman, M.M., 2018, A review on the use of polyethylene terephthalate (PET) as aggregates in concrete, *Malays. J. Sci.*, 37 (2), 118–136.

[3] Keresztes, S., Tatár, E., Mihucz, V.G., Virág, I., Majdik, C., and Záray, G., 2009, Leaching of antimony from polyethylene terephthalate (PET) bottles into mineral water, *Sci. Total Environ.*, 407 (16), 4731–4735.

[4] Alam, O., Wang, S., and Lu, W., 2018, Heavy metals dispersion during thermal treatment of plastic bags and its recovery, *J. Environ. Manage.*, 212, 367–374.

[5] Chu, J., Hu, X., Kong, L., Wang, N., Zhang, S., He, M., Ouyang, W., Liu, X., and Lin, C., 2021, Dynamic flow and pollution of antimony from polyethylene terephthalate (PET) fibers in China, *Sci. Total Environ.*, 771, 144643.

[6] Kiyataka, P.H.M., Marangoni Júnior, L., Brito, A.C.A., and Pallone, J.A.L., 2024, Migration of antimony from polyethylene terephthalate bottles to mineral water: Comparison between test conditions proposed by Brazil and the European Union, *J. Food Compos. Anal.*, 126, 105859.

[7] Filella, M., 2020, Antimony and PET bottles: Checking facts, *Chemosphere*, 261, 127732.

[8] Mohammed, E., Mohammed, T., and Mohammed, A., 2017, Optimization of an acid digestion procedure for the determination of Hg, As, Sb, Pb and Cd in fish muscle tissue, *MethodsX*, 4, 513–523.

[9] Enders, A., and Lehmann, J., 2012, Comparison of wet-digestion and dry-ashing methods for total elemental analysis of biochar, *Commun. Soil Sci. Plant Anal.*, 43 (7), 1042–1052.

[10] Song, Y., Zhao, T., He, L., Zhao, Z., and Liu, X., 2019, A promising approach for directly extracting lithium from α-spodumene by alkaline digestion and precipitation as phosphate, *Hydrometallurgy*, 189, 105141.

[11] Faridah, D.N., Solihat, I., and Yuliana, N.D., 2020, Validation of mineral (Fe, Zn, and Cu) analysis methods in carbohydrate, protein and fat-rich samples using microwave digestion method, *Indones. J. Chem.*, 20 (2), 348–359.

[12] Lehtimäki, E., and Väisänen, A., 2017, Determination of metal concentrations in certified plastic reference materials after small-size autoclave and microwave-assisted digestion followed with inductively coupled plasma optical emission spectrometry, *Spectrochim. Acta, Part B*, 127, 64–69.

[13] Cerveira, C., Hermann, P.R.S., Pereira, J.S.F., Pozebon, D., Mesko, M.F., and Moraes, D.P., 2020, Evaluation of microwave-assisted ultraviolet digestion method for rice and wheat for subsequent spectrometric determination of As, Cd, Hg and Pb, *J. Food Compos. Anal.*, 92, 103585.

[14] Barela, P.S., Souza, J.P., Pereira, J.S.F., Marques, J.C., Müller, E.I., and Moraes, D.P., 2018, Development of a microwave-assisted ultraviolet digestion method for biodiesel and subsequent trace elements determination by SF-ICP-MS, *J. Anal. At. Spectrom.*, 33 (6), 1049–1056.

[15] Talio, M.C., Feresin, V., Muñoz, V., Acosta, M., and Fernandez, L.P., 2019, New analytical methodology for Sb(III) traces quantification as emergent contaminant in drinks packaged PET samples by solid surface fluorescence, *Am. J. Anal. Chem.*, 10 (9), 377–393.

[16] Nour, A.H., Oluwaseun, A.R., Nour, A.H., Omer, M.S., and Ahmed, N., 2021, "Microwave-Assisted Extraction of Bioactive Compounds (Review)" in *Microwave Heating - Electromagnetic Fields Causing Thermal and Non-Thermal Effects*, Eds. Churyumov, G.I., IntechOpen, Rijeka, Croatia.

[17] Manzoor, M.F., Hussain, A., Sameen, A., Sahar, A., Khan, S., Siddique, R., Aadil, R.M., and Xu, B., 2021, Novel extraction, rapid assessment and bioavailability improvement of quercetin: A review, *Ultrason. Sonochem.*, 78, 105686.

[18] Tran, Q.T., Vu Thi, T.L., Do, T.L., Pham Thi, H.M., Hoang Thi, B., Chu, Q.T., Lai Phuong, P.T., Do, H.N., Hoang Than, H.T., Ta Thi, T.T., Luu, V.H., Mai Duong, P.T., and Thu Phung, H.T., 2020, Optimization of microwave-assisted extraction process of *Callicarpa candicans* (Burm. f.) Hochr essential oil and its inhibitory properties against some bacteria and cancer cell lines, *Processes*, 8 (2), 173.

[19] Goodlaxson, B., Curtzwiler, G., and Vorst, K., 2017, Evaluation of methods for determining heavy metal content in polyethylene terephthalate food packaging, *J. Plast. Film Sheeting*, 34 (2), 119–139.

[20] Othman, N.Z., Mohamad Hanapi, N.S., Saim, N., Wan Ibrahim, W.N., and Anis, A.L., 2020, Selective determination of acidic drugs in water samples using online solid phase extraction liquid chromatography with alginate incorporated multi-walled carbon nanotubes as extraction sorbent, *Indones. J. Chem.*, 20 (5), 987–999.

[21] Fan, G., Han, Y., Gu, Z., and Chen, D., 2008, Optimizing conditions for anthocyanins extraction from purple sweet potato using response surface methodology (RSM), *LWT-Food Sci. Technol.*, 41 (1), 155–160.

[22] Hocking, R.R., 2013, *Methods and Applications of Linear Models: Regression and the Analysis of Variance*, 3rd Ed., John Wiley & Sons, Hoboken, NJ, US.

[23] Bezerra, M.A., Santelli, R.E., Oliveira, E.P., Villar, L.S., and Escaleira, L.A., 2008, Response surface methodology (RSM) as a tool for optimization in analytical chemistry, *Talanta*, 76 (5), 965–977.

[24] Kainthola, J., Kalamdhad, A.S., and Goud, V.V., 2020, Optimization of process parameters for accelerated methane yield from anaerobic co-digestion of rice straw and food waste, *Renewable Energy*, 149, 1352–1359.

[25] Karlsson, T., Forsgren, C., and Steenari, B.M., 2018, Recovery of antimony: A laboratory study on the thermal decomposition and carbothermal reduction of Sb(III), Bi(III), Zn(II) oxides, and antimony compounds from metal oxide varistors, *J. Sustainable Metall.*, 4 (2), 194–204.

[26] Rastogi, L., Ankam, D.P., Yadlapalli, S., and Dash, K., 2021, Development of microwave assisted-UV digestion using diluted reagents for the determination of total nitrogen in cereals by ion chromatography, *Curr. Res. Food Sci.*, 4, 421–428.

[27] Gouda, P., 2012, *Arsenic, Selenium, Antimony Ultra-trace Analysis*, iUniverse, Bloomington, IN, US.

[28] Cibilakshmi, G., and Jegan, J., 2020, A DOE approach to optimize the strength properties of concrete incorporated with different ratios of PVA fibre and nano-Fe_2O_3, *Adv. Compos. Lett.*, 29, 2633366X20913882.

[29] Taleuzzaman, M., 2018, Limit of blank (LOB), limit of detection (LOD), and limit of quantification (LOQ), *Org. Med. Chem. Int. J.*, 7 (5), 127–131.

[30] López, A.R., Binda, G., Roncoroni, G., Recchia, S., Monticelli, D., and Spanu, D., 2024, Optimizing antimony speciation analysis via frontal

chromatography–ICP-MS to explore the release of PET additives, *Molecules*, 29 (12), 2870.

[31] Zmit, B., and Belhaneche-Bensemra, N., 2019, Antimony leaching from PET plastic into bottled water in Algerian market, *Environ. Monit. Assess.*, 191 (12), 749.

[32] Kiyataka, P.H.M., Dantas, S.T., Albino, A.C., and Pallone, J.A.L., 2018, Antimony assessment in PET bottles for soft drink, *Food Anal. Methods*, 11 (1), 1–9.

[33] Limbeck, A., Bonta, M., and Nischkauer, W., 2017, Improvements in the direct analysis of advanced materials using ICP-based measurement techniques, *J. Anal. At. Spectrom.*, 32 (2), 212–232.

[34] Magana-Maldonado, L.M., Wrobel, K., Espinoza Cruz, T.L., Yanez Barrientos, E., Corrales Escobosa, A.R., and Wrobel, K., 2023, Application of hydride generation - microwave plasma - atomic emission spectrometry and partial least squares regression for the determination of antimony directly in water and in PET after alkaline methanolysis, *Chemosphere*, 313, 137316.

[35] Capra, L., Manolache, M., Ion, I., and Ion, A.C., 2016, Validation of a method for determination of antimony in drinking water by ICP-OES, *U.P.B. Sci. Bull., Ser. B*, 78 (3), 103–112.

[36] Snedeker, S.M., 2014, "Antimony in Food Contact Materials and Household Plastics: Uses, Exposure, and Health Risk Considerations" in *Toxicants in Food Packaging and Household Plastics: Exposure and Health Risks to Consumers*, Springer London, London, UK, 205–230.

[37] Fu, X., Xie, X., Charlet, L., and He, J., 2023, A review on distribution, biogeochemistry of antimony in water and its environmental risk, *J. Hydrol.*, 625, 130043.