

Dielectric Property of Hexagonal Boron Nitride-Filled Epoxy Coatings Electrophoretically Deposited by Various N-Methylethanolamine-Cationized Epoxy-hBN Baths and EPD Voltages

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DOI: <https://doi.org/10.47772/IJRISS.2026.100900059>

Received: 08 August 2026; Accepted: 14 August 2026; Published: 30 September 2026

ABSTRACT

This study explores the dielectric property of hexagonal boron nitride-filled epoxy coatings electrophoretically deposited by various N-Methylethanolamine-cationized epoxy-h-BN baths and EPD voltages. The cationic epoxy suspension was synthesized using diglycidyl ether of bisphenol and MEA as main reactants, followed by mechanical mixing with poly(diallyldimethylammonium chloride)-functionalised h-BN particles. The electrophoretic deposition behaviour (i.e. deposit yield, coating thickness) and the coating's dielectric constant were investigated by varying MEA volumes (0.5, 1.0, and 1.5 mL) and EPD voltages (30, 40, and 60 V). The coated samples were characterised using field-emission scanning electron microscopy (FESEM), energy-dispersive X-ray spectroscopy (EDS), and electrochemical impedance spectroscopy (EIS) to evaluate morphology, elemental composition, and dielectric performance. The findings indicate optimal coating properties was achieved using 0.5 mL MEA EPD bath and deposited at 30 V, which shows a balance between deposition yield, uniformity, and dielectric behaviour. The study provides insights into the optimization of EPD-based epoxy-hBN coatings for industrial applications requiring advanced electrical insulation and protective functionalities.

Keywords: Cationic DGEBA, epoxy resin, electrophoretic deposition, dielectric properties, hexagonal boron nitride, composite coatings.

INTRODUCTION

The increasing demand for advanced materials with superior electrical and thermal properties has led to the exploration of various composite systems. Among these, epoxy resins have emerged as a widely utilized class of polymers due to their excellent mechanical strength, adhesion, and dielectric properties (Dallaev et al., 2023; Verma et al., 2025; Wang et al., 2024). However, their performance can be further enhanced through the incorporation of fillers, such as hexagonal boron nitride (h-BN) (Koay et al., 2026; Ullah et al., 2026; Wang et al., 2021). This material has gained considerable attention due to its unique two-dimensional (2D) structure and excellent thermal conductivity, coupled with outstanding electrical insulation characteristics (Ghosh et al., 2024; Naclerio & Kidambi, 2022).

In recent years, researchers have shown significant interest in developing epoxy composites filled with h-BN

to create coatings that exhibit improved dielectric properties suitable for various applications, including the electronics, aerospace, and automotive sectors (Fathi et al., 2022; Shen et al., 2021; Wan et al., 2024). The efficacy of these composites largely depends on the dispersion of h-BN within the epoxy matrix and the interfacial interactions between the filler and the polymer (Dallaev et al., 2023; Shen et al., 2021; Song et al., 2021). In order to optimize materials usage while catering to complex surface designs, electrodeposition coating methods such as electrophoretic deposition (EPD), present a promising avenue for achieving composite coating formation at an industrial scale. EPD is a versatile and cost-effective technique for fabricating uniform coatings with enhanced protective and dielectric properties, thanks to its highly tunable deposition kinetics (Shen et al., 2021). However, achieving coating uniformity in terms of thickness and material distribution requires the optimization of multiples variables that control the EPD process.

In this study, we investigate the dielectric properties of h-BN-filled epoxy coatings electrophoretically deposited using various N-Methylethanolamine (MEA)-cationized epoxy-hBN baths and EPD voltages. Optimization of the concentration of the MEA-cationized epoxy and the h-BN particle suspension is important to facilitate better interaction with the h-BN particles, which leads to a more effective and homogeneous coating during the EPD process. To achieve this, we investigate how various parameters, such as EPD voltage and bath composition, affect the dielectric constant of the resultant coatings. In particular, we examine the influence of MEA volume on deposition yield, coating thickness, and dielectric performance, providing insights into optimizing epoxy-hBN composite coatings for enhanced functionality.

This research not only seeks to advance the understanding of the dielectric behaviour of these composite coatings but also aims to contribute to the development of more effective materials for applications requiring excellent electrical insulation combined with enhanced thermal performance. Through rigorous characterization of the dielectric properties of the synthesized coatings, we hope to lay a foundation for future innovations in the field of dielectric materials, with particular emphasis on their practical applicability in real-world scenarios. Ultimately, this work will provide insights into the potential of h-BN-filled epoxy coatings as next-generation dielectric materials owing to their unique compositional advantages and processing methodologies.

METHODOLOGY

EPD Bath Preparation

A commercially available DGEBA, identified as Auto-Fix 8800-A from Chemibond Enterprise, was used as the starting material for the cationisation reaction process. The as-received DGEBA liquid has a density of 1.13 g/mL, an epoxide equivalent weight (EEW) of 198–205 g/eq, and a dynamic viscosity ranging from 500 to 1,000 centipoise (Chemibond, 2025). Cationic DGEBA epoxy resin suspensions were synthesized by a chemical reaction with six reagent-grade chemicals, including MEA, as detailed in Table 1. The formulations of the chemical reactants and the heating profile were adapted from the methodologies described by Bosso et al. (1974) and Wismer et al. (1983).

Table 1 Chemical reactants and amount to synthesise cationic epoxy resin for EPD bath

No	Chemical Reactant	Quantity
1	Epoxy Resin	140 mL
2	Dimethylether of diethylene glycol	20 mL
3	1- Octanol	41 mL
4	Stannous Chloride	1.5 grammes
5	N-Methylethanolamine (MEA)	0.5, 1.0, and 1.5 mL
6	Formic acid	0.1 mL
7	Deionized water	60 mL

Initially, the DGEBA liquid and other components (items 2-4) were mechanically stirred in a jacketed glass reactor, which was heated by a connected heating circulator. The mixture was maintained at a temperature of 130°C for 2 hours and 50 minutes under an inert environment, created by a consistent nitrogen gas flow (10

mL/min, 99.9% purity) into the reactor, which removed the air and minimized oxidation and reduced unwanted reactions. Despite being initially immiscible, the mixture became homogeneous with continued stirring and heating. After this period, the mixture was cooled at 70°C-92°C before gradually adding MEA and the remaining chemicals (items 6-7 in Table 1). The volume of MEA varied from 0.5 to 1.5 mL to produce three cationic epoxy suspensions. MEA served as a cationisation agent to generate a cationic DGEBA suspension. After adding all components, the heating circulator was turned off to commence the natural cooling process, which took approximately 2-3 hours. Mechanical stirring and nitrogen flow continued until the cationic epoxy suspension reached room temperature. Finally, 0.6 wt% of PDDA-functionalised h-BN powders (of the epoxy basis) were added to the 75 mL cationic epoxy suspensions and then hand stirred and sonication for 45 minutes. Prior to the mixing, the PDDA-functionalised h-BN was prepared by dispersing the raw h-BN particles into a poly(diallyldimethylammonium chloride) (PDDA) solution and then the PDDA-functionalised h-BN particles were retrieved using centrifugal and decantation processes. The prepared cationic epoxy-h-BN suspensions were then labelled as 0.5 mL, 1.0 mL and 1.5 mL MEA EPD baths (refer Table 2).

Electrophoretic Deposition and Curing Processes

The EPD setup includes a DC power supply (model E3643A, with voltage range of 0–35 V or 0–60 V and current ranges of 1.4 A or 0.8 A), a digital multimeter (DMM, model 34465A), four probe wires, Keysight software for real-time monitoring of DMM readings, and a beaker containing 25 mL of the EPD bath of cationic epoxy-hBN particles suspension. Commercially available galvanised steel plates (dimensions of 26×40×0.14 mm) were cleaned and then used as anode and cathode. Both electrodes are arranged in a parallel orientation with a 10 mm separation and then submerged 30 mm deep into the assigned EPD bath. Before deposition process, each of the EPD baths was ultrasonicated for 15 minutes to create a well-dispersed suspension. Each EPD process was performed using the assigned EPD bath (see Table 2) at a constant DC EPD voltage for a time duration of 15 minutes. Each of the corresponding EPD process produces one epoxy-hBN coated steel sample (retrieved from the cathode after EPD). All these samples were cured in a dry oven at 100°C for 24 hours before characterisations.

Table 2 List of EPD baths and corresponding EPD voltages used during EPD process

EPD Bath No.	EPD Bath Labeling	MEA Volume (mL) used during cationization of epoxy	h-BN loading (mg) added during cationic epoxy-h-BN mixing	EPD Voltage (V) applied during EPD process
1	0.5 mL MEA	0.5	150	30
2	0.5 mL MEA	0.5	150	40
3	0.5 mL MEA	0.5	150	60
4	1.0 mL MEA	1.0	150	30
5	1.0 mL MEA	1.0	150	40
6	1.0 mL MEA	1.5	150	60
7	1.5 mL MEA	1.5	150	30
8	1.5 mL MEA	1.5	150	40
9	1.5 mL MEA	1.5	150	60

Characterisation Methods

All characterisations of the cured epoxy coatings were carried out under standard room temperature and pressure conditions. Deposit mass of cured epoxy-hBN coating was obtained using a digital weighing balance (resolution: 0.001 grams, Explorer) by subtracting the mass of the blank cathode from the mass of the coated cathode after curing process. After that, deposit yield was calculated by dividing deposit mass with deposited coating area. The surface morphologies of the epoxy coating samples were analysed using field-emission scanning electron microscopy (FESEM) and Energy Dispersive X-ray Spectroscopy (EDS) with a Hitachi High-Tech SU5000 instrument. Prior to the FESEM-EDS characterisations, the coating samples were sputtered coated with a thin layer of gold-palladium (Au-Pd) to ensure high resolution FESEM imaging. Electrochemical Impedance Spectroscopy (EIS) of the coating was performed to evaluate the dielectric properties of the cured epoxy coating using a Gamry Instruments Reference 600 potentiostat.

The EIS samples were cut from a rectangular-shaped coated sample into an 18 mm diameter discs, and then sandwiched between two stainless-steel electrodes in a test cell. EIS measurements were taken across alternating voltage's frequencies ranging from 1 to 100,000 Hz. The real (Z') and imaginary (Z'') impedance data obtained from the EIS measurements were converted into dielectric constants (E') using the method described by Joshi et al. (2017). The EIS set-up was based on methodologies outlined by Koh et al. (2021). To check the coating thickness uniformity, cross-sectional optical micrographs of the EIS samples were obtained using Leica DVM6 at 300 $\times$ magnification. Finally, the coatings' thickness was measured at five different locations on the micrograph using LAS X software.

RESULTS AND DISCUSSION

The results demonstrate that the MEA volume and applied voltage significantly influence the deposit yield, coating thickness, elemental composition, and dielectric behaviour of the epoxy-hBN coatings. Deposit yield data illustrated in Fig. 1 represents the deposition of epoxy-hBN coating on the whole depositing surface, whereas the coating thickness data showed in the same figure, exhibits the deposition thickness at the coating's centre region. At EPD voltage of 30 V, both the deposit yield and coating thickness decreases with the increasing MEA volume, thus both properties are coherent in terms mass-thickness relationship.

However, the deposit yield and coating thickness trends at higher EPD voltages (i.e. 40 and 60 V) and higher MEA volume (i.e. 1.0 and 1.5 mL) suggest the corresponding coating thickness and material uniformity became increasing inconsistent along the depositing surface and also among the different MEA-cationised EPD baths.

Optical micrographs of Fig. 2 illustrate that coatings deposited at lower MEA volumes and EPD voltages exhibit a more uniform coating thickness, whereas those at higher MEA volumes and EPD voltages display decreased in thickness consistency. Furthermore, higher measured coating thickness is shown when EPD baths with 0.5 mL MEA volume were used, with higher EPD voltage using the 0.5 mL MEA EPD bath did not lead to significantly thicker coating. Furthermore, the noticeable white-coloured region demonstrates the presence of hBN particles inside the epoxy coating. Non-uniform h-BN distribution is demonstrated on the coating deposited using 60 V and 0.5 mL MEA.

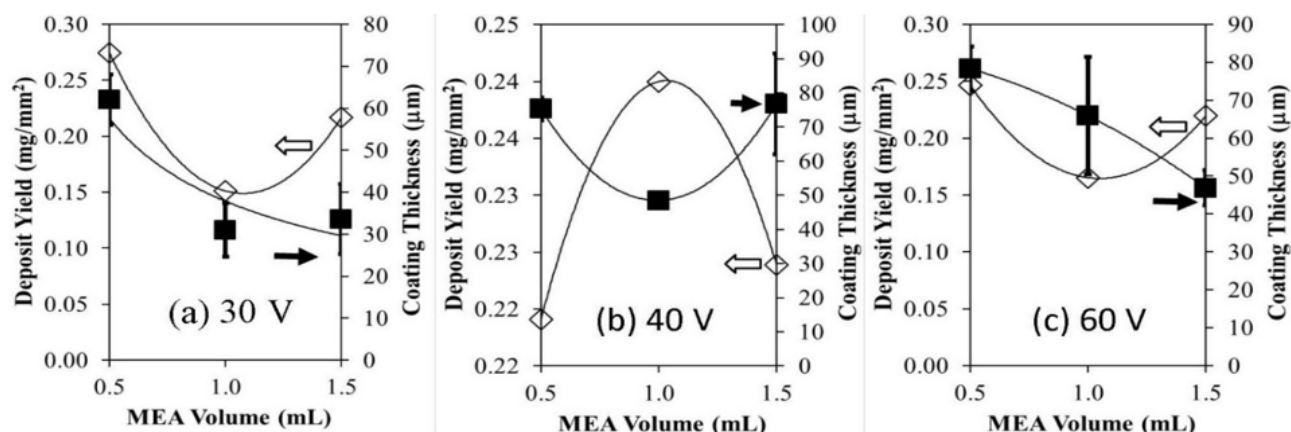


Fig. 1 Deposit yield and coating thickness versus increasing MEA volumes and EPD voltages

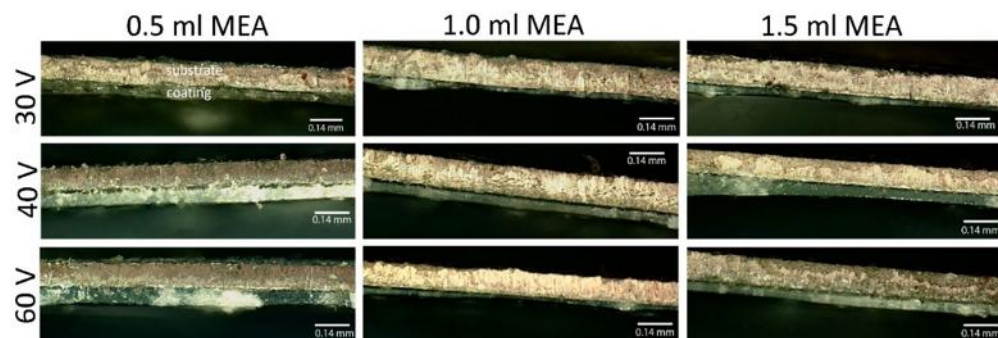


Fig. 2 Optical micrographs show the thickness of the epoxy-h-BN coating deposited using different MEA volumes and EPD voltages

EDS analysis as shown by Table 3 revealed variations in boron, carbon, nitrogen, and oxygen elements in the deposited epoxy-hBN coatings across different MEA volumes and EPD voltages. At 30 V, the boron content rose from 5.9 at% to 10.9 at% as the MEA volume reduced from 1.5 to 0.5 mL, while at 60 V, it increased from 7.6 to 14.5 at% in the 0.5 mL MEA coating sample. These changes suggest that lower MEA volume and higher EPD voltages facilitate increased deposition of boron-rich h-BN particles. As higher boron content was detected, a higher nitrogen content was also detected. Theoretically, the nitrogen content comes from the hBN particles and MEA; therefore, it should be equal to or higher than the boron content. It is possible the boron content data has larger error than the nitrogen content data due to the lower peak intensity of the former (see Fig. 3(c)). The detected oxygen content in the epoxy-hBN coating appears lower than that in the epoxy coating deposited using the pure cationic epoxy EPD bath under the same EPD parameters (Mohd Halim et al., 2025).

Table 3 Elemental data of the cured epoxy-hBN coating obtained using different EPD baths (i.e. different MEA volumes) and EPD voltages

EPD (V)	Voltage	MEA (mL)	Element (at%)			
			Boron	Carbon	Nitrogen	Oxygen
30		0.5	10.9	75.4	7.2	6.5
		1.0	5.9	84.1	2.0	8.0
		1.5	7.1	83.3	2.4	7.2
40		0.5	5.9	84.7	1.4	8.0
		1.0	6.6	83.5	2.7	7.1
		1.5	6.7	84.3	1.8	7.3
60		0.5	14.5	70.6	8.9	6.0
		1.0	6.1	84.1	1.9	7.9
		1.5	7.6	83.1	2.1	7.2

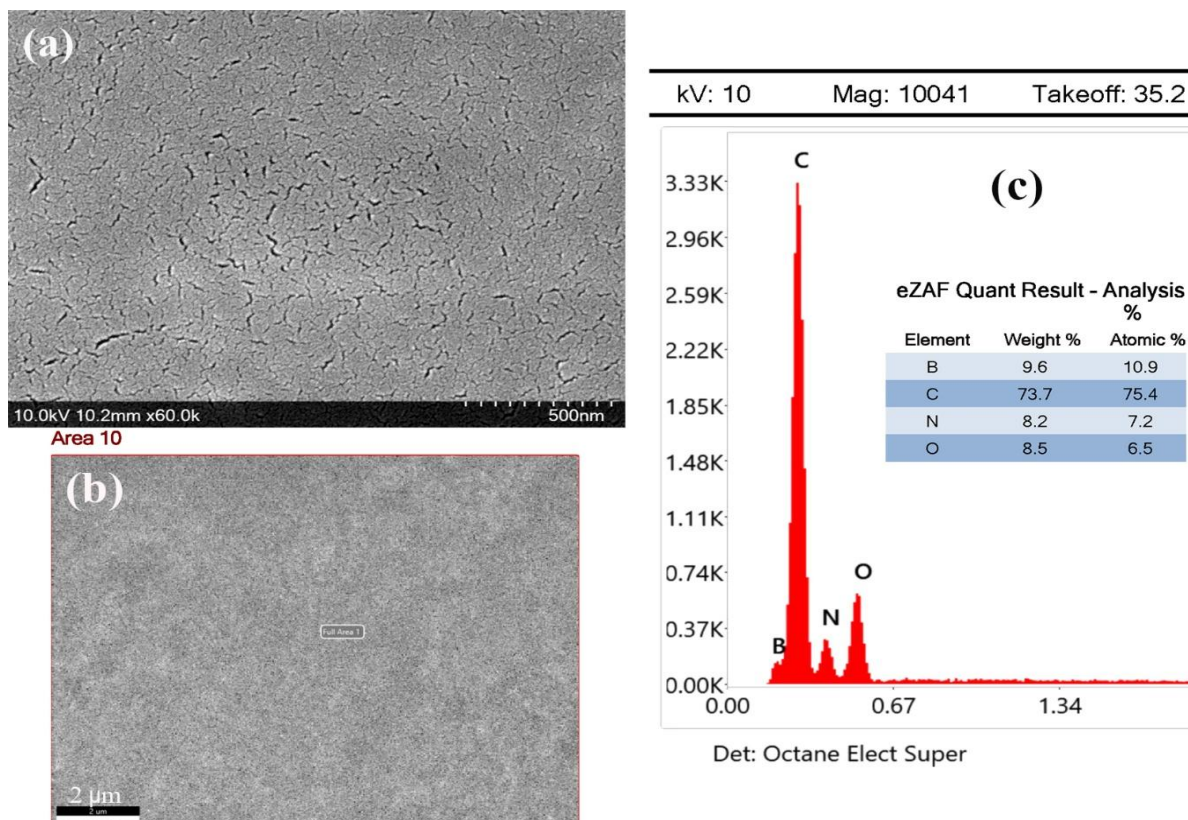


Fig. 3 (a) FESEM micrograph of planar coating surface of epoxy-hBN coating deposited using 0.5 mL MEA EPD bath and 30 V, (b) corresponding EDS scanning area of 10 μm × 12 μm, and (c) EDS spectrum and the generated elemental counts

Dielectric analysis by electrochemical impedance spectroscopy (EIS) as illustrated in Fig. 4 confirmed that coatings obtained at 0.5 mL MEA and 30 V and 40 V exhibited higher dielectric constant values at high EIS

voltage's frequency. At low EIS voltage's frequency and EPD voltage of 30 and 60 V, the dielectric constant of the 1.5 mL MEA coating displayed superior dielectric constant. This suggests that optimised MEA volume and voltage contribute to enhanced dielectric property, making these coatings suitable for protective and electronic applications. The noncorrelated relationship between the EIS and deposit yield and thickness data imply the dielectric constant of the coating sample was also determined by other factor such as the coating's microstructure. Nevertheless, further study and characterization need to be conducted to verify the possible factor.

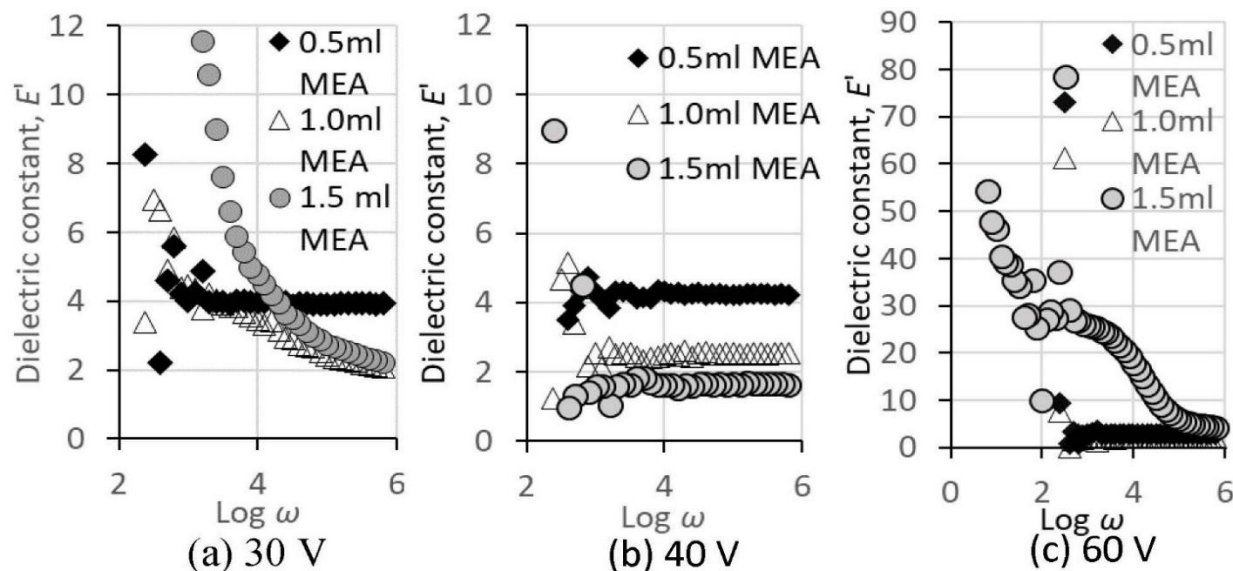


Fig. 4 Graph dielectric constant, E' vs. $\log \omega$ of epoxy coatings obtained using different EPD bath solutions (i.e. different MEA volumes) and EPD voltages. ω is the angular frequency of applied EIS voltage

CONCLUSION AND FUTURE RESEARCH

This study highlights the significant influence of MEA volume and deposition voltage on the coating thickness, composition, and dielectric property of electrophoretically deposited epoxy-hBN coatings. The results demonstrate that EPD using EPD bath derived using lower MEA volume (0.5 mL) enhance h-BN particles deposition, improving dielectric property throughout the studied EIS voltage's frequency range. However, higher MEA volumes (1.0 and 1.5 mL) result in non-uniform coatings with inconsistent thickness and a lower h-BN particles deposition. Optimal coating properties was achieved using 0.5 mL MEA EPD bath and at EPD voltage of 30 V, which provides a balance between deposition yield, uniformity, and dielectric behaviour. These findings contribute to the advancement of EPD-based epoxy coatings with tailored protective and functional characteristics, making them suitable for industrial applications requiring superior electrical insulation and mechanical durability. Further research should explore the long-term stability and environmental resistance of these coatings to expand their applicability.

ACKNOWLEDGEMENT

The authors are grateful to Universiti Teknikal Malaysia Melaka for the facilities support, particularly to Dr Wan Mohd Farid Wan Mohamad for the usage of EIS equipment. Special thanks to the Ministry of Higher Education for financially supporting this research under the Fundamental Research Grant Scheme (FRGS) research grant numbered FRGS/1/2022/TK10/UTEM/02.

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