Development of Green Composite Film for Food Paper Packaging Intercalated with Turmeric Oil



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ABSTRACT: The applications of petroleum-derived packaging materials, compared to edible coatings derived from biopolymer-based sources, lack inherent antimicrobial and barrier properties. In this study, a green-composite film for food packaging applications using turmeric oil intercalated to form biopolymer composite was developed. The bio-composite solution was prepared via the sol-gel process, utilizing a mixture of corn starch, carboxymethyl cellulose (CMC), chitosan, glycerin, and turmeric oil. The biopolymer composite was applied to unkrafted paper (UP) using roll-coating method and characterized for tensile strength (TS), surface morphology, thermal stability, surface wettability, and antimicrobial activity. The results were compared between UP and krafted paper coatings (KPCs). KPC revealed a decrease in the TS from 38.9 to 20.55 MPa and WCA also increased from 89.42 to 106° while water content and absorption increased from 7.42 to 29.32% and declined from 54.06 to 20.42%, respectively. The results also showed that both paper samples exhibited different levels of antimicrobial activity. The antimicrobial properties of the novel composite material were evaluated against *Pseudomonas aeruginosa* (PA), *Staphylococcus aureus* (SA), and *Candida albicans* (C.A) using disc diffusion method. The results showed that both coated paper samples demonstrated clear zones of inhibition for all tested microorganisms, confirming its antimicrobial efficacy. The developed green-composite film exhibited improved mechanical and antimicrobial properties, making it a promising material for food packaging applications.

KEYWORDS: Green Composite, Thin-film, Kraft Paper, Food Packaging, Turmeric Oil, Antimicrobial Materials

[Received Jan. 12, 2025; Revised Feb. 18, 2025; Accepted Mar. 2, 2025] Print ISSN: 0189-9546 | Online ISSN: 2437-2110

I. INTRODUCTION

Green packaging based on biodegradable composite materials has currently gained great attention in many food industries because of its unique properties when compared to classical petrochemical-based plastics (Pinto et al., 2021; Hussani et al., 2024). The urgent shift towards sustainable practices necessitates replacing plastic with given eco-friendly alternatives, the devastating environmental and health consequences of plastic pollution. A prime culprit is packaging, which has emerged as a critical focal point in promoting sustainable food consumption (Guillard et al., 2018). The disposal of food products inevitably results in the abandonment of packaging, exacerbating ecological pressures. To mitigate this, there is a pressing need to transition from petroleumderived packaging materials to those derived from renewable sources (Van Hai et al., 2020).

Biopolymers is one of the emerging food packaging materials with a lot of advantages over petroleum-derived packaging materials, such as biocompatibility, biodegradability, eco-friendly nature and non-toxicity (Perera *et al.*, 2023). Presence of barrier properties contribute to food quality and enhanced shelf-life of food products (Horst *et al.*, 2020). Some draw backs such as moisture, gases, aroma and lipids exacerbate longtime food storage, which inhibit microbial growth can lead to

loss of taste, spoilage and ultimately economic loss (Hou *et al.*, 2019).

Application of biopolymers have been used in different studies. Arezoo et al., (2020) incorporated starchcinnamon oil with TiO₂ nanoparticles to enhance oxygen, mechanical strength, and water vapor permeability and as well as antimicrobial capabilities for potential active food packaging of fresh pistachio. The results reveal that tensile strengths (TS) were 18 and 25 MPa for starch (5%) and $TiO_2(3\%)$. Similarly, alginate was prepared with aloe vera and garlic oil to improve the thermal and mechanical properties for the edible coating material of tomatoes. The performance shows that 100°C mass loss was recorded at the first stage, followed 218 and 266°C in the second and third, respectively. However, the tensile strength was obtained alginate (2%), aloe vera (5%) and garlic oil at 17, and 21.85 MPa respectively ((Abdel Aziz and Salama, 2021). In another work, pectin was intercalated with copaiba oil nano-emulsions to improve elongation at break and antimicrobial activity for active food packaging. The elongation break reveals pectin and copaiba were 1.7 and 2.4 MPa and with an improved TS (Norcino et al., 2020). Studied by Xin et al., 2020, zein-potato starch-chitosan nanoparticles was incorporated with curcumin. The investigation allows for high oxidation resistance and relative release efficiency of this materials on the shelf-life up to 15 days. The elongation at break were 19.1 and

50.3% for zein-potato starch and zein-potato starchchitosan with curcumin and an increase in the tensile strength from 7.9 to 13.1%.

The objective of this study is to investigate the effects of biopolymer material intercalated with turmeric oil on unkrafted paper (UP). In this study, biopolymer materials were used to prepare bio-composite for the coating of unkrafted paper. Characterizations such as tensile strength, scanning electron morphology (SEM), Fourier-transform infrared spectroscopy (FTIR), contact angle (CA) analysis, and thermal gravimetric analysis (TGA) of both unkrafted paper (UP) and krafted paper (KP) coated with biocomposites were carried out. The wettability and antimicrobial activity were conducted on the papers.

II. MATERIALS AND METHOD

A. Materials

Cassava starch and carboxymethyl cellulose (CMC) obtained from Muda Lawal market, Bauchi, Nigeria, were utilized in this study. Chitosan was purchased from GHK, Mainland, China. Turmeric oil was supplied by Like Sun GmbH, Planck, Germany. Glycerin was purchased from Green Herbology, Puchong, Selangor, Malaysia. White oil was obtained from USP Phama Grade, Malaysia. Unbleached brown kraft paper was obtained from central market, Bauchi, Nigeria. *Pseudomonas aeruginosa (ATCC 27853), Staphylococcus aureus (ATCC 25923), and Candida albicans (ATCC 10231)* microbial strains for estimating the antimicrobial activity of the paper samples

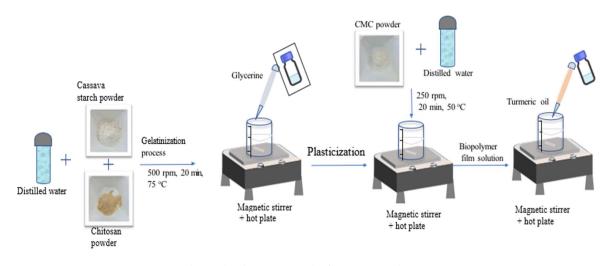
Were obtained from the Microbiological Culture Collection Center in the Faculty of Science, Universiti Teknologi Malaysia. The selection of these bacterial and fungal strains was based on their potential application in enhancing the shelf life of food packaging. All materials were used as received.

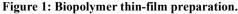
B. Method

The development of bio-composite-based coating materials involved preparing coating solutions using the sol-gel method, followed by the application of a thin-film coating solution to unbleached kraft paper. Finally, the coated papers underwent a post-treatment drying process.

Preparation of bio-composite thin film coating solution

The development of a biopolymer coating for unkrafted paper (UP), illustrated in Fig.1. The gelatinous process was carried out by dissolving 1.32 g of cassava starch and 1.65 g of chitosan powder in 37.25 mL of distilled water at 75°C with stirring for 20 minutes. Then, 2.10 g of glycerin was added dropwise to the solution. Next, 0.1 g of CMC powder was dispersed and thoroughly mixed in distilled water for 20 minutes at 50°C, after which the two solutions were combined and continuously stirred to form a homogeneous biopolymer film solution. Finally, 4.0 g of turmeric oil was added to the solution and stirred continuously.





Coating process of unkraft paper (UP)

The unbleached brown paper sheet (unkrafted paper: UP) (10 cm x 10 cm) was coated with the prepared bio-polymer thin film solution using a glass rod. A pipette was used to dispense 2.5 mL of the solution onto the paper's front spot. The coated paper

was then rolled up and dried at 60°C for 15 minutes as indicated in Fig.2. This coating process was repeated for four cycles, yielding four differently coated paper samples: KPC1, KPC2, and KPC3.

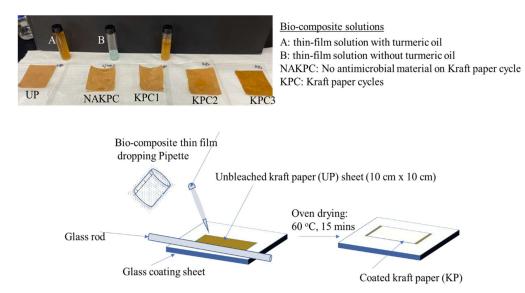


Figure 2: Coating process on unkraft paper (UP) using biopolymer thin-film solution and number of coating cycles (C1-3).

III. CHARACTERIZATION

A. Tensile strength

Tensile strength tests were conducted on unkrafted paper (UP) and krafted paper coatings (KPC) samples using a tensile tester. The objective was to evaluate the effect of bio-composite coatings on the tensile strength of paper. Tests were conducted using a grip-grip separation strength device (ZwickRoell GmbH & KG, D-89079 Ulm, Germany). The tests employed a 0.5 N pre-load cell, 50.0 mm initial equipment separation, and 200 mm/min initial speed. Paper samples were placed in between the slit of a 50 mm x 50 mm sample holder/gripper, with width determined from SEM images. The testing procedure followed the ASTM D 828-16 standard for tensile strength test (paper).

B. Morphological Analysis

SEM analysis (Hitachi TM3000) examined the UP and KPC surface and cross-section. Samples (1.0 cm) were gold/platinum-coated (5 min) before imaging. To prevent charging, the samples were coated with gold using an argon plasma metallizer (sputter coater K575X, Edwards Limited, UK). SEM imaging was conducted at an acceleration voltage of 3 kV and at different magnifications.

C. FTIR Analysis

Fourier transform infrared spectroscopy (FTIR) analysis was performed to identify and compare the functional groups present on the UP and KPC samples. Using a PE Frontier FTIR (model number 96064) spectrometer, the adsorption spectra were recorded over a wavenumber range of 650-4000 cm⁻¹ with a 8 cm⁻¹ resolution in transmittance mode.

D. TGA Analysis

Thermogravimetric analysis (TGA) was performed using a TA Instrument Q-500 to investigate the thermal properties of UP and KPC samples. The analysis employed a step-wise isothermal method, where samples were heated from 38° C to 800° C under airflow at a heating rate of 5° C/min.

E. Water Content Analysis

Water content was measured according to ASTM D 644. Samples were dried at 105 °C for 24 h, and weight loss was calculated using Eq. (1). Average water content was determined from 3 replicates.

Water content(%) =
$$\frac{MM_{ff} - MM_i}{MM_{ff}}$$
 100 (1)

F. Water Adsorption

Water absorption was measured according to ASTM D 570. Samples were dried (50 °C, 24 h), cooled in a desiccator, and then immersed in distilled water (RT, 6 h). Initial (M) and final (MM_{ff}) masses were recorded, and data were evaluated using Eq. (2).

Water adsorption(%) =
$$\frac{MM_{ff} - MM_i}{MM_{ff}}$$
 100 (2)

G. Contact Angle Analysis

Contact angle measurements (OCA 15 EC, Dataphysics) assessed the wettability in terms of their hydrophobicity and oleophobicity of UP and KPC. The sessile drop method involved dispensing 0.5 μ L of deionized water and 0.1 μ L of different oil (glycerin, white and vegetable oil) samples, capturing high-resolution images, and calculating average contact angles from repeated measurements.

H. Antimicrobial Activity Evaluation

A modified agar diffusion method (Alebooyeh *et al.*, 2012; Arezoo *et al.*, 2020) was employed to evaluate the antibacterial and antifungal properties of UP and KPC1, KPC2 and KPC3. The composite-coated kraft paper discs were prepared and placed on Mueller-Hinton agar inoculated with 0.2 mL of

IV. RESULTS AND DISCUSSION

SEM analysis of the UP (Figs.3[ai-aii]) reveals a smooth, porous surface with a random arrangement of fibers. At 1000x magnification, the fibers appear to be 45.4 μ m in diameter, with visible twists and kinks. The surface exhibits moderate roughness, with valleys and ridges spaced approximately 5-10 μ m apart. Pores and

bacterial suspensions (approximately 105–106 CFU/mL). The plates were incubated at 37°C for 24 hours for bacteria and 30°C for 48 hours for the fungal strain. Meanwhile, 5 mm diameter film disks were exposed to UV light for 30 minutes before being placed on the agar plates. Following incubation, the presence of zones of inhibition around the test discs of the UP and KPC samples were recorded.

voids, ranging from 1-5 μ m in diameter, are scattered throughout the surface. The fiber bundles are loosely packed, leaving noticeable gaps between them. However, the analysis of the KPC (Figs.3[bi-bii]) reveals a relatively uniform, 35.1 μ m thick coating layer.

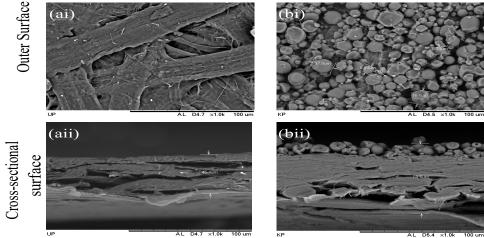


Figure 3: SEM images of unkrafted paper (a) and krafted paper (b).

The FTIR spectrum of UP reveals characteristic absorption peaks indicative of its chemical composition. Broad O-H stretching vibrations (3400-3200 cm⁻¹) and C-H stretching vibrations (2900-2800 cm⁻¹) signify the presence of cellulose and hemicellulose. Carbonyl groups (1740-1720 cm⁻¹) and aromatic rings (1650-1630 cm⁻¹) suggest lignin content, while ether groups (1250-1200 cm⁻¹) and alcohol groups (1030-1000 cm⁻¹) indicate cellulose and hemicellulose. These functional groups imply hydrophilicity, oxidative degradation, and lignin presence, influencing paper properties such as strength, durability, and printability (Ruggero *et al.*, 2020). FTIR analysis of KPC with bio-composites

(CMC, cassava starch, glycerin, chitosan, and turmeric oil) revealed enhanced functional group absorption, indicating successful incorporation. The spectrum as shown in Fig.4 revealed an increase O-H (3400-3200 cm⁻¹), C-H (2900-2800 cm⁻¹), C=O (1740-1720 cm⁻¹), and C-O (1250-1200 cm⁻¹) in stretching vibrations, and aromatic C=C (1650-1630 cm⁻¹) from turmeric oil, suggesting improved hydrophobicity, mechanical strength, barrier properties, and antioxidant activity. This enhancement is attributed to the hydroxyl groups from CMC, cassava starch, and glycerin, carbonyl groups from CMC and chitosan, and amide I/II bands from chitosan, implying potential applications in improved paper products (Mustapha *et al.*, 2018).

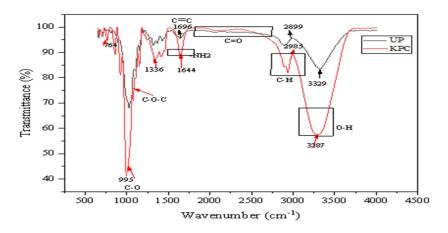


Figure 4: FTIR analysis of unkrafted paper and krafted paper.

The TGA curves of UP and KPC exhibit distinct thermal degradation patterns (Fig.5(a&b)). UP shows a single-stage degradation between 250-400°C, attributed to cellulose and hemicellulose decomposition. In contrast, KPC displays a multi-stage degradation profile, with initial weight loss (100-200°C) due to moisture and glycerin evaporation, followed by degradation of cassava starch and CMC (200-300°C), and finally, chitosan and cellulose decomposition (300-400°C).

The presence of turmeric oil is evident from the slight shift in degradation temperature and residual weight. The enhanced thermal stability of KPC, as indicated by high residual weight (5.0%) at 500°C as compared to UP higher residual weight (7.0%), suggests improved durability and resistance to thermal degradation, making it a promising sustainable material (Ruggero *et al.*, 2020)(Shah *et al.*, 2024).

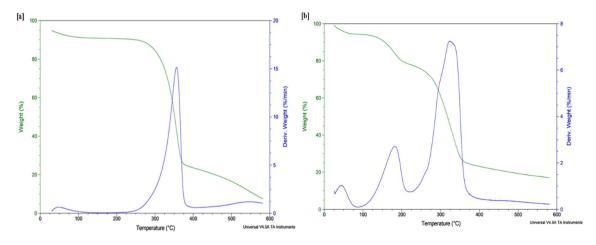


Figure 5: TGA analysis of [a] unkrafted paper (UP), [b] krafted paper coated (KPC).

The data suggests that the coating increases the water content of KP from 7.42% to 29.32%, indicating the coating's hygroscopic nature (Fig.6). The weight loss after oven drying supports this: UP lost 7.1% (44.867-41.767mg) of its initial weight, whereas KPC lost 22.7% (63.77-49.3mg). This significant difference implies the coating retains more moisture, potentially

may affect paper durability and shelf life. The 21.9% increase in water content and 15.6% greater weight loss in coated paper are statistically significant and practically relevant, indicating potential issues with packaging applications. The coating's moisture-absorbing properties should be considered when selecting materials for specific uses.

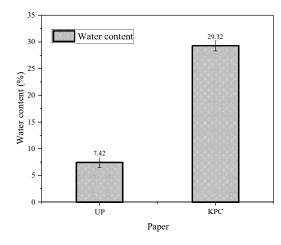


Figure 6: Water content of unkrafted paper and krafted paper.

The water absorption results reveal distinct differences between UP and KPC. UP exhibits high water absorption (mean: 54.06%, range: 70.58%) indicating suitability for dry goods packaging or low-humidity environments (Fig.7). In contrast, KPC shows relatively low water absorption (mean: 20.42%, range: 18.46%), making it suitable for packaging food,

or products requiring moisture protection. Notably, KPC absorbs significantly less water than UP, highlighting the coating's effectiveness in reducing moisture penetration. These findings suggest that coating kraft paper substantially enhances its water resistance, expanding its potential applications in packaging industries.

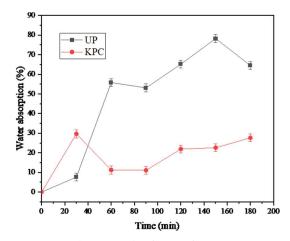


Figure 7: Water absorption analysis of unkrafted paper and krafted paper.

The tensile strength results of UP and KPC with bio-composite materials revealed varying degrees of mechanical enhancement. Notably, KPC1 showed a 1.8% increase in tensile strength (38.9 MPa) compared to UP (38.2 MPa), indicating that the bio-composite coating can enhance paper durability. However, KPC2 and KPC3 demonstrated significant decreases in tensile strength (23.3 and 20.55 respectively) (Fig.8), suggesting that variations in bio-composite Formulation or coating process affected paper integrity. These findings underscore the importance of optimizing bio-composite formulation and coating processes to achieve desired mechanical properties. The decreased tensile strength in KPC2-3 may be attributed to factors such as inadequate coating uniformity, insufficient curing time, incompatible bio-composite ratios, or inadequate adhesion between coating and paper (Liu *et al.*, 2018).

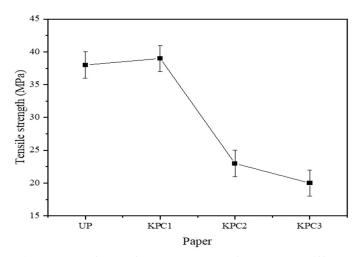


Figure 8: Tensile strength of unkrafted paper and krafted paper at difference coating cycles.

The water contact angle (WCA) and oil contact angle (OCA) measurements revealed variations in hydrophobicity significant and oleophobicity between uncoated unrafted paper (UP) and coated Kraft paper (KPC) samples. The UP exhibited a WCA of 75.3° and OCA of 100°, indicating moderate hydrophobicity and oleophobicity. In contrast, KPC samples demonstrated enhanced hydrophobicity, with WCA values ranging from 89.42° (KPC1) to 110° (KPC3). Notably, KPC2displayed WCAs of 103.03°. The bio-composite coatings' effectiveness in improving hydrophobicity and oleophobicity is evident, particularly with KPC3 showing the highest WCA. The OCA values remained relatively consistent across KPC samples, indicating stable oleophobic properties. These findings suggest that the bio-composite coatings, comprising CMC, cassava starch, turmeric oil, glycerin, and chitosan,

successfully enhanced the water and oil repellency of KPC samples. The variations in WCA among KPC samples may be attributed to differences in coating thickness, composition, or curing conditions. Further optimization of the bio-composite formulation and coating process could lead to even more significant improvements in hydrophobicity and oleophobicity. In addition, CA measurements revealed varying degrees of oleophobicity on KPC for different oils. Vegetable oil exhibited a relatively low contact angle of $31.3^{\circ} \pm$ 24.93°, indicating poor oleophobicity (Fig.9). In moderate white oil demonstrated contrast. oleophobicity with a contact angle of $82.77^{\circ} \pm 80.05^{\circ}$. Notably, glycerine showed the highest contact angle of $106.93^{\circ} \pm 103.77^{\circ}$, indicating excellent oleophobicity and resistance to wetting. These results highlight the KPC's selective oleophobic properties, making it suitable for applications such as food packaging.



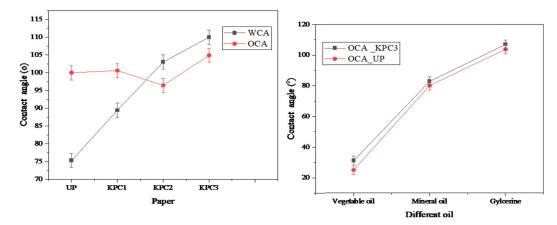


Figure 9: Contact angles of unkrafted paper and krafted paper at difference coating cycles.

The antimicrobial activity of the uncoated and kraft paper samples against Pseudomonas aeruginosa, Staphylococcus, and *Candida Albicans* was evaluated. The results showed that the KP sample exhibited a higher zone of inhibition (ZOI) against *Pseudomonas aeruginosa* (2-2.3 cm) compared to the kraft paper sample (0 cm). In contrast, the KPC sample showed a higher ZOI against *S. aureus* (1-3 cm) compared to the uncoated paper sample (0 cm). The antifungal activity of both paper samples was comparable, with ZOI measurements ranging from 2.2-2.7 cm (Fig.10). KPC showed excellent antimicrobial activity against *P*.

aeruginosa and S. aureus as compared to C. Albicans.

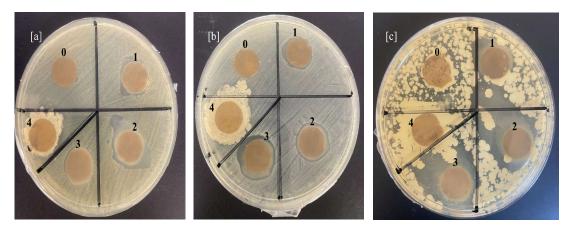


Figure 10: Antibacterial activity test on UP, NAKPC:0, KP1:1, KP2:2 and KP3:3 by disk diffusion method *(a) S. aureus;* for S.A S- 0 cm zone with bacterial growth UP- 0 cm zone, KP1- 2 cm zone, KP2- 3 cm zone KP3- 1.5 cm zone. (b) *P. aeruginosa;* For P.A S- 0 cm zone with bacterial growth UP- 0 cm zone KP1- 1.8 cm zone KP2- 2 cm zone KP3- 2.3 cm zone and *(c) C. Albicans;* for fungi (C.A) S- 0 cm zone with fungal growth UP- 0 cm zone KP1- 2.2 cm zone, KP2- 2.7 cm zone, KP3- 3 cm zone.

V. CONCLUSION

This study developed a green-composite film for food packaging applications using turmeric oil intercalated between unkraft paper (UP) and a biopolymer composite. The composite was formulated using corn starch, CMC, chitosan, glycerin, and turmeric oil. The biopolymer composite was applied to unkraft paper and characterized for tensile strength, morphology, thermal stability. surface and antimicrobial properties. The results were compared to kraft paper to evaluate the suitability of unkraft paper as a sustainable alternative. The developed greencomposite film exhibited improved mechanical and antimicrobial properties, making it a promising material for food packaging applications. The study also evaluated the antimicrobial activity of uncoated and kraft paper samples against P. aeruginosa, S. aureus. and antifungal (Candida Albicans) microorganisms. KPC showed excellent antimicrobial activity against P. aeruginosa and S. aureus as compared to C. Albicans with no resistance against UP. Future studies should investigate coating thickness and uniformity effects, optimize bio-composite ratios and curing conditions, and explore alternative biocomposite materials. The successful development of bio-composite-coated Kraft paper with enhanced tensile strength has potential applications in sustainable packaging, paperboard, and specialty papers, aligning with the growing demand for ecofriendly materials.

AUTHOR CONTRIBUTIONS

Y. O. Raji: Writing, Editing, Supervision, Fund acquisition, Conceptualization, J. Mohammed: Fund acquisition, A. Salis: Conceptualization, A. Mohammed: Conceptualization, M. S. Muhammad: Writing, Data collection

ACKNOWLEDGEMENTS

The authors gratefully thank the financial support from the Tertiary Education Trust Fund under the institution-based research (IBR) Grant Scheme (TETF/DR&D/CE/UNI/BAUCHI/IBR/2022/VOL.1)

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