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The Effect of Spent Coffee Grounds (SCG) as a Natural Filler on the Physio Mechanical Properties of Polyvinyl Alcohol (PVA) Films

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ABSTRACT. Polyvinyl alcohol (PVA) is a biodegradable polymer that is safe, colorless, odorless, and non-toxic. It forms strong, flexible, and translucent films, making it highly suitable for various applications such as packaging, adhesives, and coatings. However, despite its excellent swelling capacity, PVA is highly sensitive to water due to the presence of hydrophilic hydroxyl groups in its side chains. This study investigates the incorporation of spent coffee grounds (SCG) as a natural filler to enhance the physio-mechanical and water-resistance properties of PVA films. PVA/SCG composite films were fabricated via solution casting using varying SCG loadings (0–2 g) and citric acid as a crosslinker. FTIR analysis confirmed the presence of characteristic PVA and lignocellulosic SCG functional groups, along with shifts in C=O and O–H regions indicating altered hydrogen-bonding environments. Tensile testing showed that increasing SCG content reduced tensile strength from 2.31 MPa (0 g SCG) to 0.63 MPa (2.0 g SCG), while Young's modulus increased due to the rigid structure of SCG. Water absorption decreased markedly with filler incorporation, dropping from 21.61% (0 g SCG) to 5.57% (2.0 g SCG), demonstrating improved water resistance. Overall, SCG serves as an effective bio-filler that enhances moisture barrier properties and stiffness of PVA films, suggesting their potential for sustainable packaging applications.

Key words: Bioplastics, natural filler, Spent coffee ground, Polyvinyl alcohol (PVA), films

1. INTRODUCTION

The escalating environmental concerns associated with non-biodegradable, petroleum-based plastics have led to a significant shift in focus toward the development of sustainable, eco-friendly materials. Biodegradable polymers, such as polyvinyl alcohol (PVA), have emerged as promising alternatives due to their excellent film forming properties, biodegradability and non-toxicity, making it suitable for various applications, including food packaging, biomedical devices, and agricultural mulch films (Hussain et al., 2023). However, despite these advantages, pure PVA films often exhibit inadequate mechanical properties, poor water resistance and a relatively high production cost, which limits their widespread application (Jamasri et al., 2023). A major contributor to PVA's water sensitivity is the abundance of hydrophilic hydroxyl groups along its molecular chains, which readily interact with water molecules, causing swelling or even dissolution in humid environments (Liu et al., 2022). Improving the water resistance of PVA without compromising biodegradability remains a central challenge. To address this issue, two complementary approaches are commonly employed. The first involves chemical crosslinking, such as using citric acid (CA), which reduces hydrophilicity by forming ester linkages that consume free hydroxyl groups. The second approach relies on physical reinforcement with natural fillers, which enhances mechanical stability and creates tortuous pathways that

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slow water diffusion. Although natural fillers such as cellulose nanocrystals, microcrystalline cellulose, and starch are themselves hydrophilic, their incorporation can still indirectly reduce water uptake by increasing matrix density, filling micro voids, and reinforcing the polymer network. Thus, the goal is not to make the fillers less hydrophilic, but to use them to create a filler-induced barrier effect that limits water permeation while also improving mechanical properties. This strategy aligns with circular economic principles by transforming agricultural and food industry into valuable functional fillers.

Spent coffee grounds (SCG) represent a globally abundant waste material rich in lignocellulosic components such as cellulose, hemicellulose, lignin, and residual lipids (McNutt & He, 2019). Prior research has examined SCG fillers in polymers such as PLA and PP, with mixed mechanical performance due to limited compatibility (Alharbi et al., 2024; Mäder et al., 2025). While some studies have used SCG-derived holocellulose or extracts in PVA matrices, the direct incorporation of unmodified, whole SCG into PVA matrices has not been thoroughly studied. Understanding how whole SCG affects hydrogen bonding network, mechanical performance and water uptake is necessary to optimize PVA based bioplastics for sustainable applications.

Therefore, this present study aims to systematically investigate the influence of increasing SCG filler concentrations on the physio-mechanical and water absorption properties of PVA films. Specifically, the objectives are to evaluate the tensile properties of PVA/SCG composite films and examine the interactions between SCG and the PVA matrix through FTIR analysis, while also considering the combined effects of SCG reinforcement and CA crosslinking on water resistance.

2. METHODOLOGY

2.1 Preparation of Spent Coffee Grounds (SCG)

SCG was collected from a local café immediately after brewing. The grounds were washed with distilled water to remove residual impurities, oven-dried at 80 °C for 24 hours, , and sieved to <250 µm to ensure uniform particle size and moisture content prior to incorporation**.**

2.2 Preparation of PVA/SCG films

PVA films incorporated with SCG fillers were prepared using a solution casting method. Initially, 2.5 g PVA was dissolved in 50 mL of distilled water by continuous heating at approximately 90 °C for 1 hour to ensure complete dissolution. Once the PVA was fully dissolved, 3 g of CA and 5 mL of glycerol were added as a crosslinking agent and plasticizer, respectively. SCG fillers (0–2 g) were incorporated and stirred magnetically for 15 minutes, to ensure unform filler dispersion. Table 1 enumerates the composition of the PVA/SCG films. The composite mixture was then maintained under heat for a further 15 minutes to ensure uniform dispersion of the fillers. Subsequently, the resulting solution was cast onto a glass plate and dried overnight in a drying cabinet at ambient temperature. The dried films were then subjected to heat treatment at 140 °C for 30 minutes to enhance crosslinking and film integrity. Film thickness was measured at five points using a micrometer. Samples were conditioned at 23 °C and 50% relative humidity for 48 hours before testing.

Table 1. Composition of the PVA/SCG composite films

Mass of PVA (g)	Mass of Citric Acid (g)	Glycerol (mL)	Mass of SCG (g)	Sample Designation
2.50	3.00	5.00	0.00	SCG 0
			0.50	SCG 0.5
			1.00	SCG 1.0
			1.50	SCG 1.5
			2.00	SCG 2.0

2.3 Characterization and Testing

2.3.1 Fourier Transform Infrared Spectroscopy (FTIR)

The spectra of the film samples were acquired at ambient temperature utilizing FTIR (Thermo Fisher Scientific Nicolet iS 10) with Attenuated Total Reflectance (ATR). The objective of the FTIR analysis was to determine the interaction between PVA and SCG. Prior to conducting transmittance mode measurements at a resolution of 2 cm^{-1} with 16 scans across the frequency range of 4000 cm^{-1} to 600 cm^{-1}, the samples were positioned directly onto the crystal.

2.3.2 Tensile Testing

The tensile properties of the film samples, including tensile strength, elongation at break and Young modulus, were assessed using the Instron Universal Testing Instrument (3365, Instron USA). Prior to testing, all film samples were conditioned at 23 ± 2 °C and 50 ± 5% relative humidity for 48 hours, in accordance with ASTM D882. Film thickness was measured at five random points using a digital micrometer (±0.001 mm), and the average value was used for mechanical property calculations. Each specimen was sectioned into 1.0 cm x 7.0 cm segments and assessed at a crosshead speed of 10 mm/min. The tests were conducted on three distinct film samples to minimize the possibility of error. The tensile test results indicate the amount of force required to break the bioplastic, as well as the extent to which the samples stretched and elongated before failure. These results provide an assessment of the films' resistance to mechanical stress.

2.3.3 Water Absorption

The water absorption test was performed in accordance with ASTM D570 standards. Samples from each ratio were prepared with dimensions of 24 mm × 10 mm. Prior to testing, the thin-film samples were dried in an oven at 80 °C. Water uptake was then measured at 120-hour intervals until weight stability was achieved. The weight gain due to water absorption was calculated and reported as a percentage.

3. RESULTS AND DISCUSSION

3.1 Structural studies of PVA/SCG bio composites films

Figure 1 represents the result of FTIR for PVA/SCG composite films with varying loadings of SCG. The most identifiable peaks of PVA without SCG (SCG 0) are those corresponding to acetate and hydroxyl groups, as reported by Patil et al. (2021). The broad band at approximately 3302 cm^{-1}, with the highest intensity, is attributed to the hydrogen-bonded O–H stretching vibration of hydroxyl groups, indicating possible hydrogen bonding interactions within the material. The C–H stretching vibration of alkyl groups appears at 2936 cm^{-1}, while the C=O stretching band, caused by residual acetate groups in PVA and interaction with CA, is observed at 1709 cm^{-1}, as noted by Huang et al. (2023). Further identifiable peaks include the bending vibration of $C–H_2$ at 1413 cm^{-1}, the C–O absorption band at 1039 cm^{-1}, and the C–C absorption peak at 850 cm^{-1}.

Upon incorporation of SCG, several noticeable changes in the FTIR spectra were observed. Tthe O–H peak shifted slightly to higher wavenumbers (3315–3332 cm^{-1}), indicating reduced PVA hydrogen bonding likely due to disruption by SCG particles and the presence of SCG hydroxyl groups altering the bonding environment. The C=O band shifted within 1703–1725 cm^{-1}, suggesting enhanced esterification reactions facilitated by citric acid in the presence of SCG. Increased intensities in the 1250–1050 cm^{-1} region confirm lignocellulosic contributions from SCG. Peaks near 830–880 cm^{-1} became more defined at higher SCG loadings, indicating aromatic structures from lignin. No new chemical bonds were observed, suggesting the interactions are primarily physical and hydrogen-bond mediated.

With the increasing SCG loading in PVA films, several noticeable changes in the FTIR spectra were observed. The O–H stretching band, initially at 3302 cm^{-1} for neat PVA, gradually shifted to higher wavenumbers (3315–3332 cm^{-1}), indicating stronger hydrogen bonding interactions contributed by the hydroxyl groups of SCG. The C–H stretching peaks also showed slight shifts from 2936 cm^{-1} (0 g SCG) to 2949 cm^{-1} (2.0 g SCG), suggesting the increasing contribution of aliphatic groups from SCG. More importantly, the carbonyl (C=O) stretching region displayed significant variation. The neat PVA/CA film exhibited a band at 1709 cm^{-1}, while films with SCG shifted within the range 1703–1725 cm^{-1}. These shifts confirm the occurrence of esterification and enhanced crosslinking as SCG content increased, consistent with stronger interactions between SCG and CA. The C–O stretching vibrations (1250–1050 cm^{-1}) also became more pronounced with higher SCG loadings, reflecting the contribution of lignocellulosic components such as hemicellulose and cellulose. Similarly, the peak near 1030 cm^{-1}, associated with C–O–H vibrations, was consistently observed, verifying the presence of polysaccharide structures from SCG. At lower wavenumbers (830–880 cm^{-1}), peaks attributed to C–H stretching vibrations of aromatic groups became clearer with increasing SCG, further confirming the incorporation of lignin-derived structures. Overall, the progressive peak shifts and intensity changes with increasing SCG loading indicate stronger interactions between SCG, PVA, and CA. These results suggest that higher SCG content enhances the complexity of the crosslinked network through hydrogen bonding and esterification, thereby modifying the structural arrangement of the films.

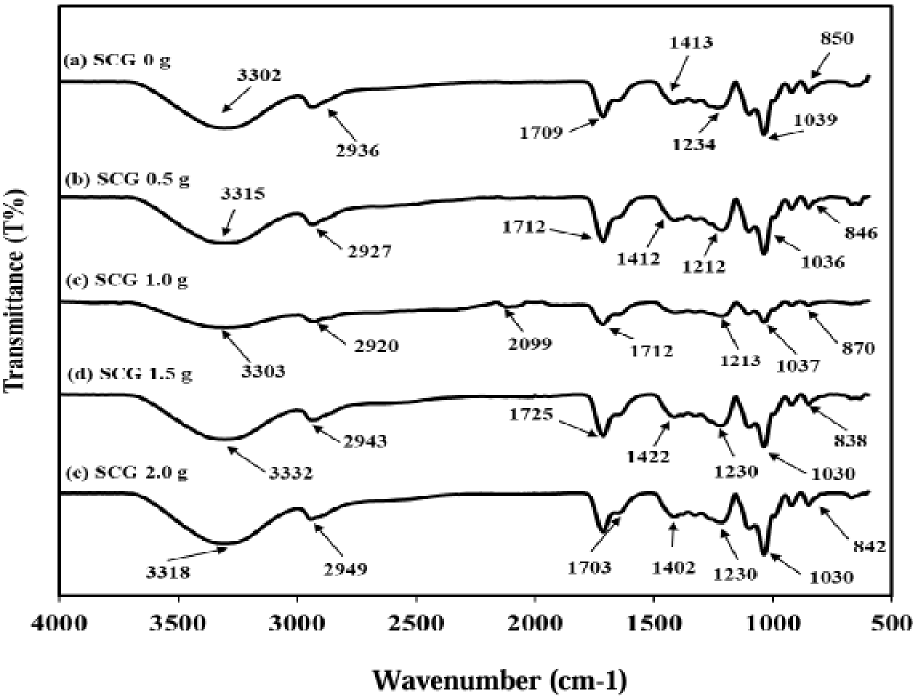

Figure 1. FTIR spectra of PVA/SCG bio composites films with varying SCG loadings

3.2. Tensile Properties of PVA/SCG bio composites films

The tensile test is a vital technique for evaluating the mechanical properties of materials, their strength and elasticity. By applying a tensile force to a specimen until failure, key parameters such as tensile strength, elongation, and Young modulus can be determined. The results of this tensile test are presented in Figures 2, 3, and 4, respectively. Tensile strength is the maximum stress a material can withstand before fracturing when subjected to stretching or pulling, providing an indication of the material's ability to resist deformation under tension (Pal et al., 2022). Based on the data presented in Figure 2, the tensile strength of pure PVA films reached 2.3107 MPa, indicating that in the absence of SCG filler, the material retained its inherent mechanical properties. However, with the incorporation of SCG filler, a progressive decline in tensile strength was observed: 1.5056 MPa at 0.5 g, 1.3879 MPa at 1.0 g, 0.9044 MPa at 1.5 g, and 0.6279 MPa at 2.0 g. This reduction can be attributed to the poor interfacial adhesion between the hydrophilic SCG fillers and the PVA matrix.

As the filler loading increased, the absence of strong bonding disrupted the continuity of the polymer matrix, thereby weakening the composite structure and lowering its ability to withstand tensile loads. Moderate filler loading (0.5–1.0 g) maintained better strength before significant deterioration at higher loadings. Furthermore, the lignocellulosic and hydrophilic nature of SCG promoted filler agglomeration, which further compromised the mechanical integrity of the films. According to Seo et al. (2023), polymers reinforced with natural fillers such as SCG often exhibit thermodynamic incompatibility, meaning the components are unable to fully coexist or interact effectively at the molecular level. This incompatibility further explains the observed deterioration in tensile strength with increasing SCG content.

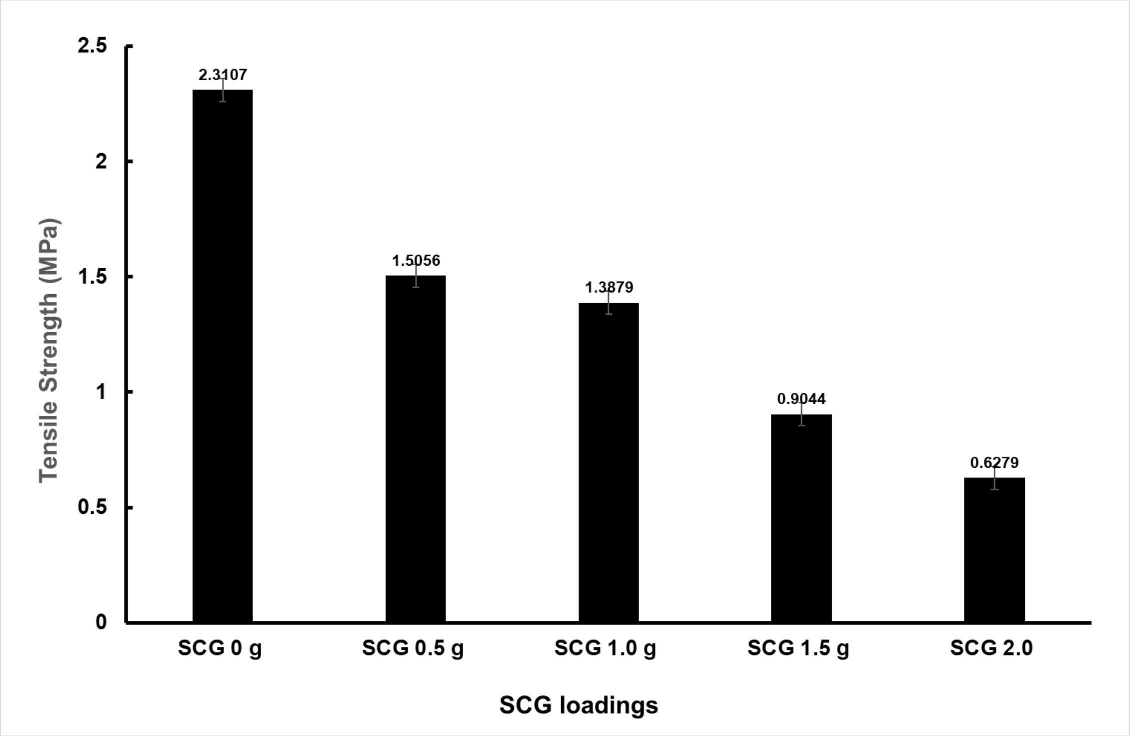

Figure 2. Tensile strength of PVA films with varying loadings of SCG filler

Elongation at break is an essential mechanical parameter that measures the highest strain a material can endure prior to failure. It is generally expressed as a percentage, representing the elongation of a material specimen relative to its initial length at the point of fracture. Figure 3 provides an overview of the percentage elongation for PVA films with varying of SCG loadings. The elongation at break of PVA films incorporating SCG filler showed a clear decreasing trend with increasing SCG content. The control PVA film recorded the highest elongation at break of 1.9328 %, while the addition of 0.5 g SCG filler caused a significant 31 % reduction to 1.3234 %, and further reductions were observed as the filler loading increased. This decline can be explained by several factors. First, the rigid SCG particles restrict the mobility of PVA molecular chains, thereby limiting the ability of the matrix to stretch before failure (Hwang et al., 2021). Second, the poor interfacial adhesion between hydrophilic SCG and the PVA matrix creates localized weak zones that act as stress concentrators, promoting earlier crack initiation during tensile deformation (Seo et al., 2023). At higher filler loadings, these stress concentrations become more pronounced, contributing to increased brittleness of the composite films. Additionally, the uneven distribution and agglomeration of SCG particles at elevated loadings likely introduce micro voids and structural defects, which serve as crack initiation sites and further diminish the elongation capacity of the films. Overall, these combined effects lead to reduced chain mobility, higher brittleness, and lower elongation at break as SCG content increases.

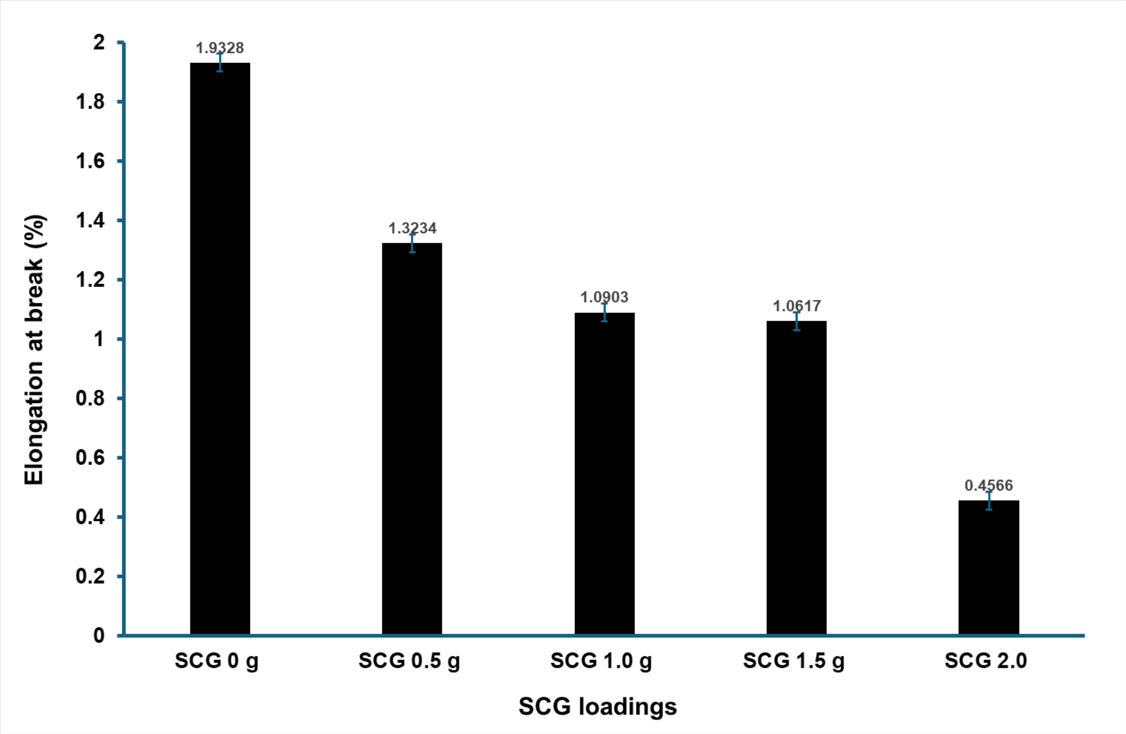

Figure 3. Elongation at break of PVA films with varying loadings of SCG filler

Young's Modulus, also known as modulus of elasticity, is a fundamental mechanical characteristic that measures a material's capacity for elastic deformation under applied stress. It is defined as the ratio of stress to strain within a material's elastic limit (Panova & Shishatskaya, 2020). Based on Figure 4, the PVA films containing SCG filler exhibited an increasing trend in Young's modulus, rising from 1.4443 MPa for pure PVA to 3.3950 MPa for the composite with the highest filler loading. This significant increase indicates that the incorporation of SCG enhances the rigidity of the composite. The improvement in stiffness can be attributed to the rigid lignocellulosic nature of SCG, which restricts PVA chain mobility and limits segmental motion within the polymer matrix (Hwang et al., 2021). The presence of rigid filler particles also creates a more constrained polymer network, reducing chain slippage when stress is applied and thereby increasing the composite's resistance to deformation. This increase in stiffness is consistent with the mechanical trends observed for tensile strength and elongation at break. While SCG addition increased the modulus, it also contributed to a reduction in tensile strength and a notable decrease in elongation, particularly at higher loadings. This indicates a trade-off between stiffness and toughness, where the films become more brittle and less capable of absorbing energy before failure. In other words, although the composite becomes stiffer with increasing SCG content, it simultaneously becomes more susceptible to fracture under stress, reflecting reduced overall flexibility and toughness at high filler concentrations.

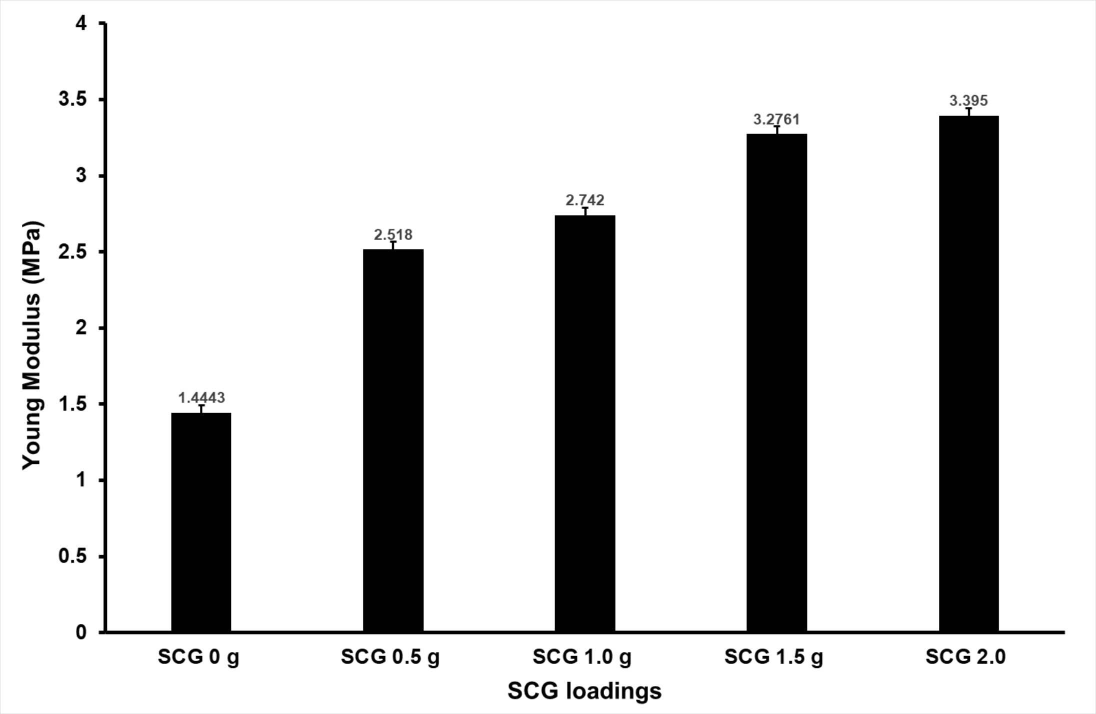

Figure 4. Young modulus of PVA films with varying loadings of SCG filler

3.3 Water Absorption

Water absorption is a critical property in evaluating the performance of polymeric films, particularly for applications involving moisture exposure. It indicates the material's ability to resist or absorb water, which can significantly impact its structural integrity, mechanical properties, and long-term durability. As shown in Figure 5, the PVA films exhibited a clear decreasing trend in water absorption with increasing SCG loading. The control sample (0 g SCG) recorded the highest water absorption at 21.61%, whereas films with added SCG showed markedly lower values, with 6.08% at 0.5 g, 3.17% at 1.0 g, 3.92% at 1.5 g, and 5.57% at 2.0 g.

Several mechanisms can explain this reduction. First, the dispersed SCG particles act as a physical barrier, occupying voids and interrupting the diffusion pathways available for water molecules. By reducing the free volume within the PVA matrix, SCG fillers increase the tortuosity of water transport, thereby slowing water penetration (Hwang et al., 2021). Second, hydrogen bonding interactions between SCG and PVA chains may reduce the number of available hydroxyl groups capable of binding water. This interaction, coupled with the presence of lignocellulosic components in SCG, potentially enhances crosslinking density in the matrix, further limiting water uptake. However, at higher filler loadings (e.g., 2.0 g SCG), slight increases in water absorption may occur due to uneven dispersion or agglomeration of SCG particles, which can introduce localized microvoids that facilitate limited water penetration. Although this effect was not dominant, it provides deeper insight into the complex interplay between filler distribution and moisture behavior. Overall, the substantial decline in water absorption demonstrates the effectiveness of SCG in enhancing the moisture resistance of PVA films. This improvement is particularly beneficial for packaging applications in humid environments, where reduced water uptake can enhance stability, protect contents, and extend material lifespan.

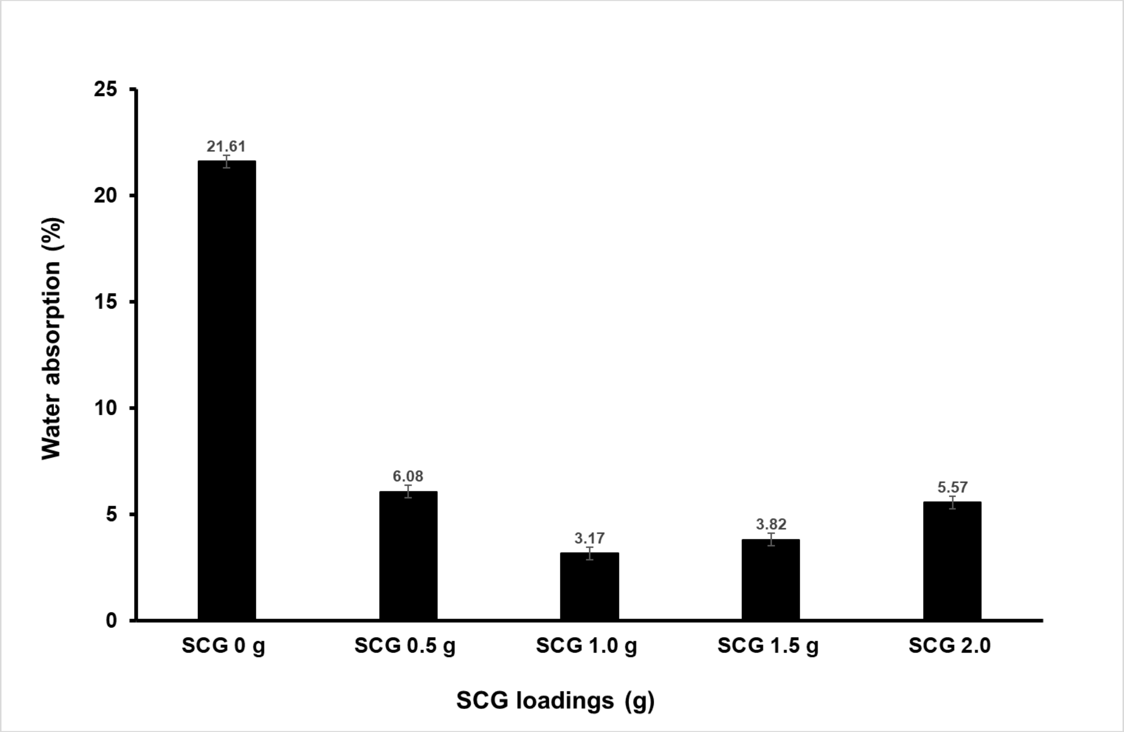

Figure 5. Water absorption of PVA films with varying loadings of SCG filler

4. CONCLUSION

The incorporation of SCG filler into PVA films significantly influenced the physio mechanical properties and moisture related properties of PVA films. Tensile strength decreased systematically from 2.31 MPa for the neat PVA film to 0.63 MPa at 2.0 g SCG, reflecting the reduced interfacial compatibility and increased brittleness with higher filler loading. In contrast, water absorption decreased substantially from 21.61% (0 g SCG) to 3.17–5.57% for SCG-reinforced films highlighting the strong barrier effect imparted by the lignocellulosic filler. Among the examined concentrations, 1.0 g SCG provided the most balanced improvement by yielding enhanced moisture resistance while retaining acceptable mechanical performance, demonstrating the importance of optimizing filler levels. FTIR analysis further supported these findings by showing no formation of new chemical bonds, indicating that the observed property changes were driven primarily by physical interactions and hydrogen-bonding modifications, rather than chemical reactions with the PVA matrix. This aligns directly with the study's objective of evaluating interfacial interactions between SCG and PVA. Overall, SCG serves as an effective natural filler capable of improving the water resistance and stiffness of PVA films, particularly at moderate loadings without introducing chemical modification. These enhancements position SCG-reinforced PVA composites as promising candidates for eco-friendly packaging applications, where moisture stability and biodegradability are essential.

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AUTHOR CONTRIBUTIONS

Mai Alifah Ilyana Mior Mohd Harith is responsible for data collection and analysis as well as writing the original draft. **Faiezah Hashim** is responsible for supervising, administrating the project and editing the final manuscript. **Siti Nor Din**, **Nur Nasulhah Kasim** and **Nabilah Akemal Muhd Zailani** are responsible for editing the final manuscript

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DECLARATION OF GENERATIVE AI IN THE WRITING PROCESS

During the preparation of this work, the author(s) used ChatGPT (OpenAI) to assist in improving the clarity and language quality of the manuscript. After using this tool/service, the author(s) reviewed, edited, and verified all content, and take full responsibility for the accuracy and integrity of the publication.

DATA AVAILABILITY

All data generated or analysed during this study are included in this published article and its supplementary information files.

COMPETING INTEREST

The authors declare that there are no competing interests.

COMPLIANCE OF ETHICAL STANDARDS

The authors declare that this research did not involve human or animal subjects and this research does not include any ethical issue. All experimental procedures were conducted in accordance with the institutional Safety, Health, and Environmental (HSE) protocols of Universiti Teknologi MARA (UiTM).

SUPPLEMENTARY MATERIAL

No supplementary material is associated with this article.

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