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Chemical treatments' effect on the structural and mechanical properties of polyvinyl alcohol/spent coffee ground composite films

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Abstract

Chemical treatments are a common strategy for improving the compatibility of natural fillers with polymer matrices. However, their specific impact on Spent Coffee Grounds (SCG) as reinforcement for Polyvinyl Alcohol (PVA) composites remains unclear. This study investigates the effects of alkalization, bleaching, and acid hydrolysis on the structural and mechanical properties of PVA/SCG composite films. The SCG was treated with 10% NaOH, 10% NaOCl, and 1 M $H_2 SO_4$. FTIR analysis showed that untreated SCG exhibited characteristic C–H stretching peaks at 2922 cm^{-1} and 2853 cm^{-1}, corresponding to methyl and methylene groups in cellulose and hemicellulose. After alkalization and bleaching, these peaks nearly disappeared, indicating excessive removal of hemicellulose and lignin and suggesting structural degradation of the filler. In contrast, acid hydrolysis largely preserved these peaks, reflecting milder structural modification. Mechanical testing confirmed this trend: the untreated composite achieved the best performance, with tensile strength of 2.30 MPa and tear resistance of 2.12 N All chemically treated samples showed reduced strength and toughness, with alkalization being the most detrimental. These findings demonstrate that the decline in mechanical properties is directly correlated with structural damage detected by FTIR, emphasizing the need to optimize treatment severity.

Keywords:
Alkalization, bleaching, acid hydrolysis, Polyvinyl Alcohol (PVA), Spent Coffee Grounds (SGC)

1 Introduction

The development of environmentally friendly materials, particularly in polymers and composites, has become a primary research focus amid growing awareness of the adverse environmental impacts of synthetic waste. Conventional polymers, which are difficult to degrade, create significant challenges in waste management, thereby driving research toward more sustainable alternatives [1]. Among these, Polyvinyl Alcohol (PVA), a synthetic polymer known for its biodegradability, flexibility, and water solubility, has garnered considerable attention. PVA's ability to interact with various natural fillers makes it a potential matrix for fabricating composite films, which have applications in diverse sectors such as packaging, biomedicine, and other green technologies [2].

Spent Coffee Grounds (SCG) are an abundant and underutilized organic waste, offering potential for development as a filler in PVA-based composite films [3]. SCG is rich in lignocellulosic compounds (cellulose, hemicellulose, and lignin) that can be incorporated into a polymer matrix. However, integrating natural fillers often faces the challenge of interfacial incompatibility. The hydrophilic nature and complex surface chemistry of SCG can affect their interaction with the PVA, which ultimately determines the final characteristics of the resulting composite material [4], [5].

To modify the characteristics of lignocellulosic fillers, various chemical treatments are often applied. Methods such as alkalization, bleaching, and acid hydrolysis are known to alter the chemical composition and surface morphology of natural materials. Alkalization is commonly used to reduce lignin and hemicellulose content, which is known to affect interfacial adhesion [6]. The bleaching process is subsequently applied to reduce color components and residual lignin, potentially altering the cellulose purity. Meanwhile, acid hydrolysis is known to break down the cellulose structure into smaller particles [7].

Chemical modification of various natural fiber-based materials has been widely investigated. Yet, there is a lack of studies specifically examining the impact of such treatments on SCG for application in PVA composite films. This research aims to systematically investigate the effects of different chemical treatments (alkalization, bleaching, and acid hydrolysis) on the mechanical properties of a PVA-based composite film containing SCG as a filler. Specifically, this study will evaluate changes in tensile strength and tear resistance to understand the relationship between filler surface modification and the material's mechanical response. The findings from this study are expected to provide fundamental insights for the utilization of organic waste, such as SCG, in the development of bio-based composites.

2 Research methodology

2.1 Materials and equipment

PVA (molecular weight 60,000 g/mol) from Sigma Aldrich, Singapore, was used as the matrix for the composite films. SCG, the filler, was obtained from a local coffee shop in Banda Aceh. Sodium hydroxide (NaOH), sodium hypochlorite (NaOCl), and sulfuric acid (H_2SO_4) were purchased from Merck (Darmstadt, Germany) and used as the chemical agents for the treatment processes.

The equipment used during the research stages included a three-neck flask, a hot plate with a magnetic stirrer, a spatula, a thermometer, an analytical balance, a furnace, an ultrafine grinder, a sieve, a centrifuge, an ultrasonic bath, a digital micrometer, and a Universal Testing Machine.

2.2 Chemical treatment

The chemical treatment method followed the approach described by Fitriani *et al.* [8]. The initial stage involved preparing SCG by washing and subsequently drying them in an oven at 60°C for 12 hours to ensure complete moisture removal. Afterward, the dried SCG were sieved through a 40 mesh. The chemical treatment processes were then carried out as follows.

a. Alkalization

The alkalization process was performed by immersing 50 grams of SCG in a 10% NaOH solution. The mixture was heated at 60°C for 45 minutes to dissolve lignin and other non-cellulosic components. Afterward, the treated SCG were washed until a neutral pH was reached and then dried in an oven at 60°C for 12 hours[9], [10], [11].

b. Bleaching

The bleaching process was conducted using a 10% NaOCl (1:1) for 1 hour at room temperature to remove color components and any residual lignin. The resulting material was then dried at 60 °C for 12 hours [6].

c. Acid Hydrolysis

Acid hydrolysis of the SCG was performed using 1 M H_2SO_4 at 45°C for 1 hour. The reaction was quenched by adding deionized water, and the mixture was left overnight to form a suspension. The suspension was washed until a neutral pH was achieved, then centrifuged at 5000 rpm for 20 minutes. This washing-centrifugation cycle was repeated 1–3 times until a suspension was obtained, which was subsequently dried [8], [12].

2.3 Film preparation

The PVA films filled with SCG were prepared by solution casting and evaporation. 10% (w/v) PVA solution was prepared by dissolving the polymer in deionized water at 80°C under continuous stirring for 15 minutes. Subsequently, the solution was degassed by cooling it in a refrigerator for 12 hours to remove any air bubbles.

The film solution was prepared by adding 5% (w/v) of SCG to the PVA solution. The mixture was stirred for 5 minutes until homogeneous and then cast into a 20x20 cm mold. The film was dried in an oven at 60°C for 12 hours and then cooled to room temperature before storage. This method was a modification of the technique developed by Alizadeh-Sani et al. [13]. The preparation of the composite film is illustrated in Fig. 1.

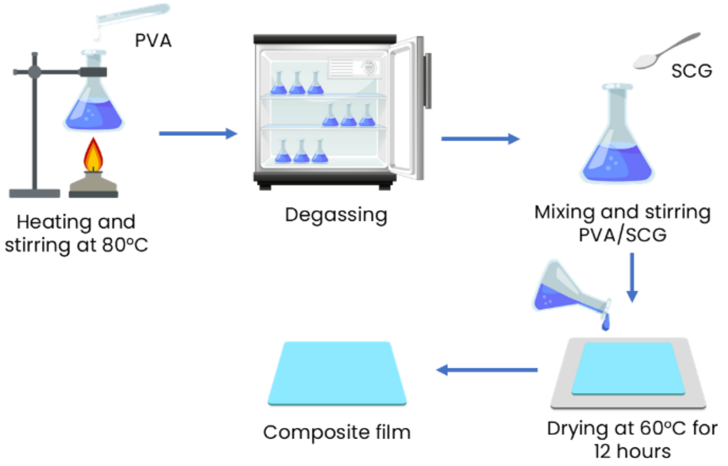

Fig. 1. Preparation of composite film

2.4 FTIR analysis

The FTIR analysis of all composite samples was performed to examine functional groups and molecular structure using a Shimadzu Prestige-6400 FTIR spectrometer. The spectra were recorded within the wavenumber range of 4000–500 cm^{-1}, following the ASTM E1252-98 standard.

2.5 Tensile testing

The tensile test specimens were prepared in a rectangular shape, measuring 110 mm in length and 20 mm in width. Of the total 110 mm length, 60 mm was designated for the grip sections, divided equally with 30 mm at the top and 30 mm at the bottom of the specimen. These dimensions were based on the ASTM D882 "Standard Test Method for Tensile Properties of Thin Plastic Sheeting." Each test was repeated 3 times [14][15]. The dimensions of the tensile test specimen are illustrated in Fig. 2.

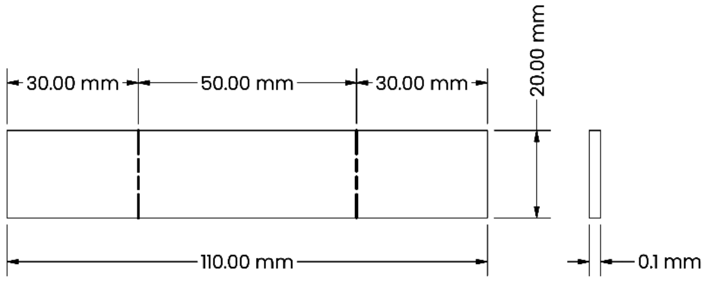

Fig. 2. Tensile test specimen

2.6 Tear test

The tear test specimen had a different shape and dimensions compared to the tensile test specimen. The specimen featured a specific 90° angle, a design intended to concentrate the applied load at its tip to initiate a tear. Each specimen was tested three times, following the ASTM D1004 "Standard Test Method for Tear Resistance (Graves Tear) of Plastic Film and Sheeting." The dimensions of the tear test specimen are illustrated in Fig. 3.

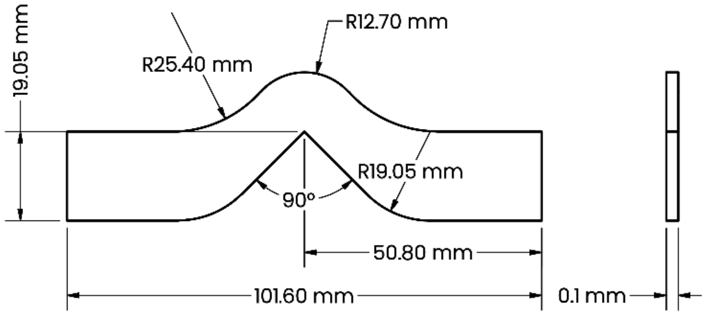

Fig. 3. Tear test specimen

3 Results and discussion

3.1 Visual appearance of composite films

A visual comparison of the four composite film samples is presented in Fig. 4. Each demonstrates the tangible impact of the chemical treatments on the SGC. Visually, the PVA/SGC and PVA/SGC-Hyd (Acid Hydrolysis) samples both retained the characteristic deep black color of the original coffee grounds. In contrast, the other chemical treatments resulted in dramatic color changes. The PVA/SGC-Alk (Alkali) sample transitioned to a uniform light brown, whereas the PVA/SGC-Ble (Bleaching) sample appeared as white.

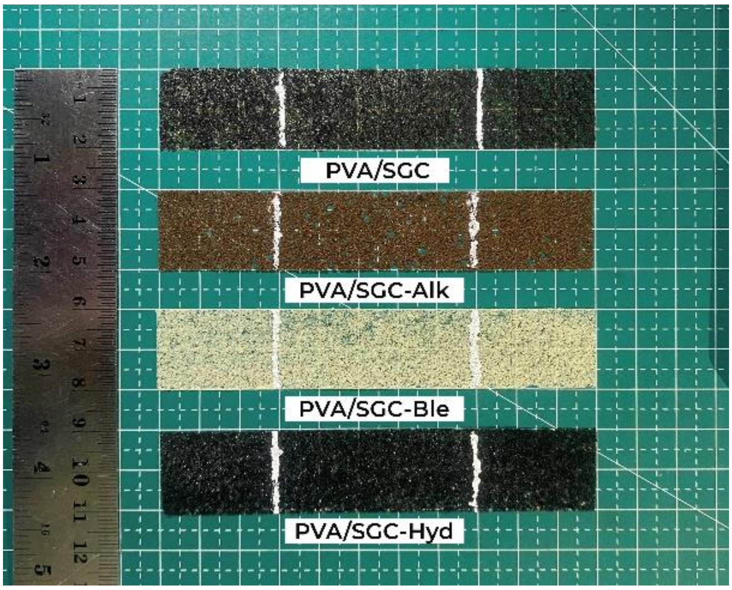

Fig. 4. Visual comparison of the four composite film samples

These color differences directly reflect the mechanism of each chemical treatment. The light brown hue of the alkali sample is attributed to the removal of a substantial portion of lignin and pigment compounds during alkalization [16]. The bleaching process took this a step further, eliminating nearly all colored components to yield cellulose of higher purity, which resulted in its pale white appearance. The fact that the hydrolysis sample remained black corroborates that this treatment is more focused on modifying the cellulose structure rather than removing pigments [17].

3.2 FTIR analysis

FTIR analysis was performed to investigate the chemical structural changes in SCG following various chemical treatments and to evaluate their implications for the composite's characteristics. The FTIR spectra of untreated SCG, as well as SCG subjected to alkalization, bleaching, and acid hydrolysis, are presented in Fig. 5.

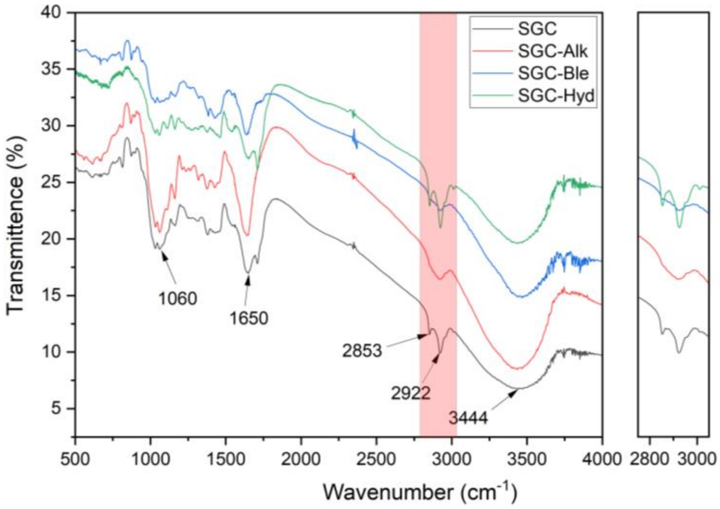

Fig. 5. FTIR spectra of SGC and the following chemical treatments

The spectrum of untreated SCG displays the characteristic peaks of a lignocellulosic material. A broad absorption band centered at 3444 cm^{-1} corresponds to the O–H stretching vibrations of hydroxyl groups in cellulose, hemicellulose, lignin, and adsorbed water [18], [19], [20]. The peaks at approximately 1650 cm^{-1} are attributed to

C=O stretching in hemicellulose and C=C aromatic skeletal vibrations in lignin, while the prominent peak at 1060 cm^{-1} is characteristic of C–O stretching in the cellulose backbone [19], [21].

Notably, the untreated SCG spectrum exhibits two distinct peaks at 2922 cm^{-1} and 2853 cm^{-1}, which correspond to the asymmetric and symmetric C–H stretching vibrations of methyl (–CH_3) and methylene (–CH_2) groups in cellulose and hemicellulose, respectively [18], [22]. However, these characteristic aliphatic C–H peaks virtually disappeared after alkalization (SCG-Alk) and bleaching (SCG-Ble), indicating an extensive removal of aliphatic-rich compounds. This disappearance suggests that both the alkali and the bleaching agent not only eliminated non-cellulosic impurities but also aggressively degraded hemicellulose and lignin, which contribute significantly to these C–H signals [23]. In contrast, acid hydrolysis (SCG-Hyd), which is more selective toward amorphous cellulose, preserved the aliphatic C–H peaks to a greater extent than the other treatments. Hence, the loss of the 2922 and 2853 cm^{-1} peaks should not be regarded as evidence of effective purification but rather as an indication of excessive treatment severity, which ultimately compromises the structural integrity of the SCG filler.

3.3 Tensile strength

The mechanical properties of the PVA/SGC, as influenced by various chemical treatments, are illustrated by the stress-strain curves in Fig. 6 and Fig. 7. The untreated PVA/SGC clearly demonstrated the superior mechanical performance. This control sample achieved the highest tensile strength, peaking at 2.30 MPa, and the greatest elongation at break, 37.5%. This behavior suggests adequate interfacial bonding between the natural coffee ground particles and the PVA polymer matrix, enabling efficient load transfer from the matrix to the filler.

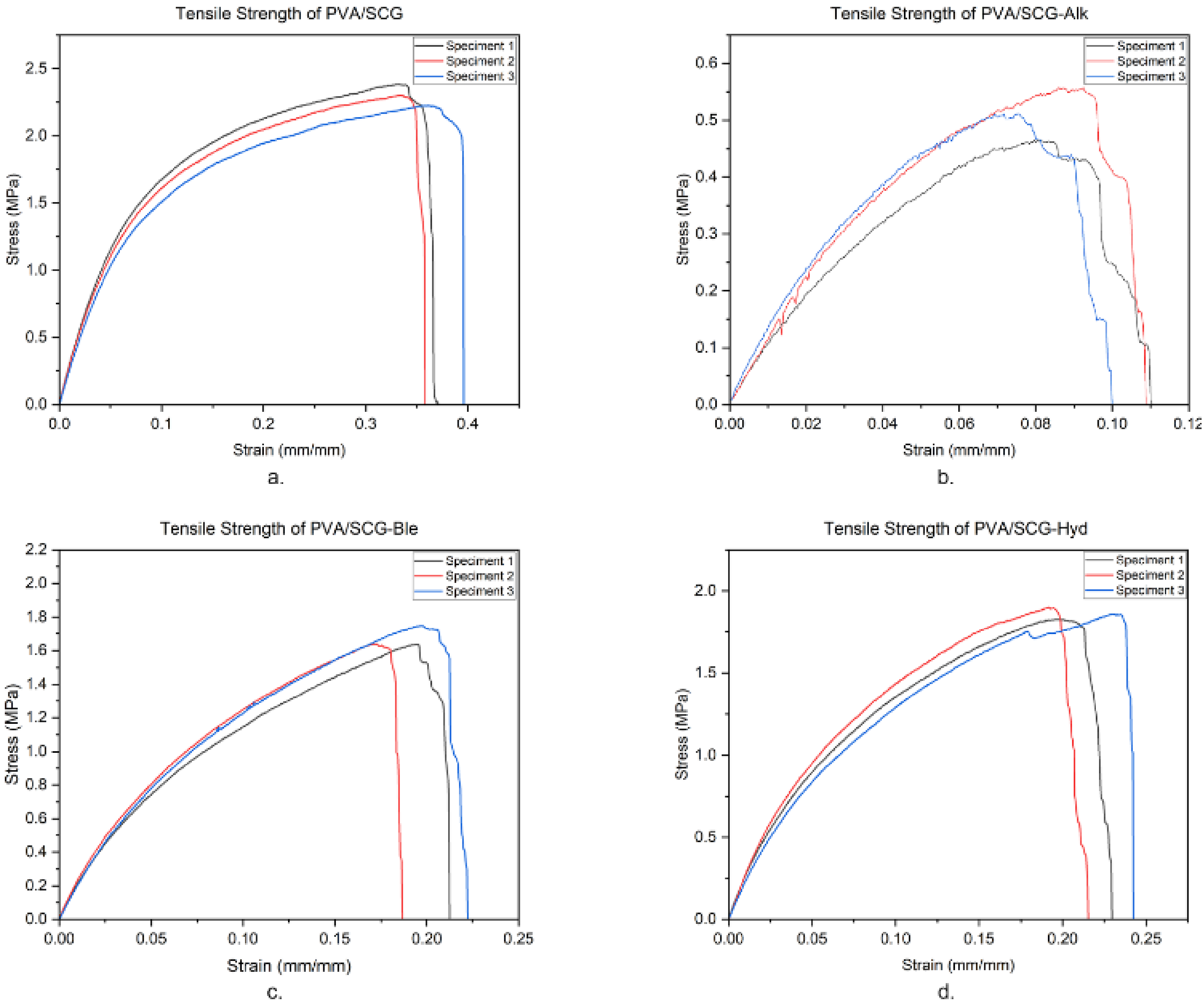

Fig. 6. Stress-strain curves: (a) PVA/SCG, (b) PVA/SCG-Alk, (c) PVA/SCG-Ble, and (d) PVA/SCG-Hyd

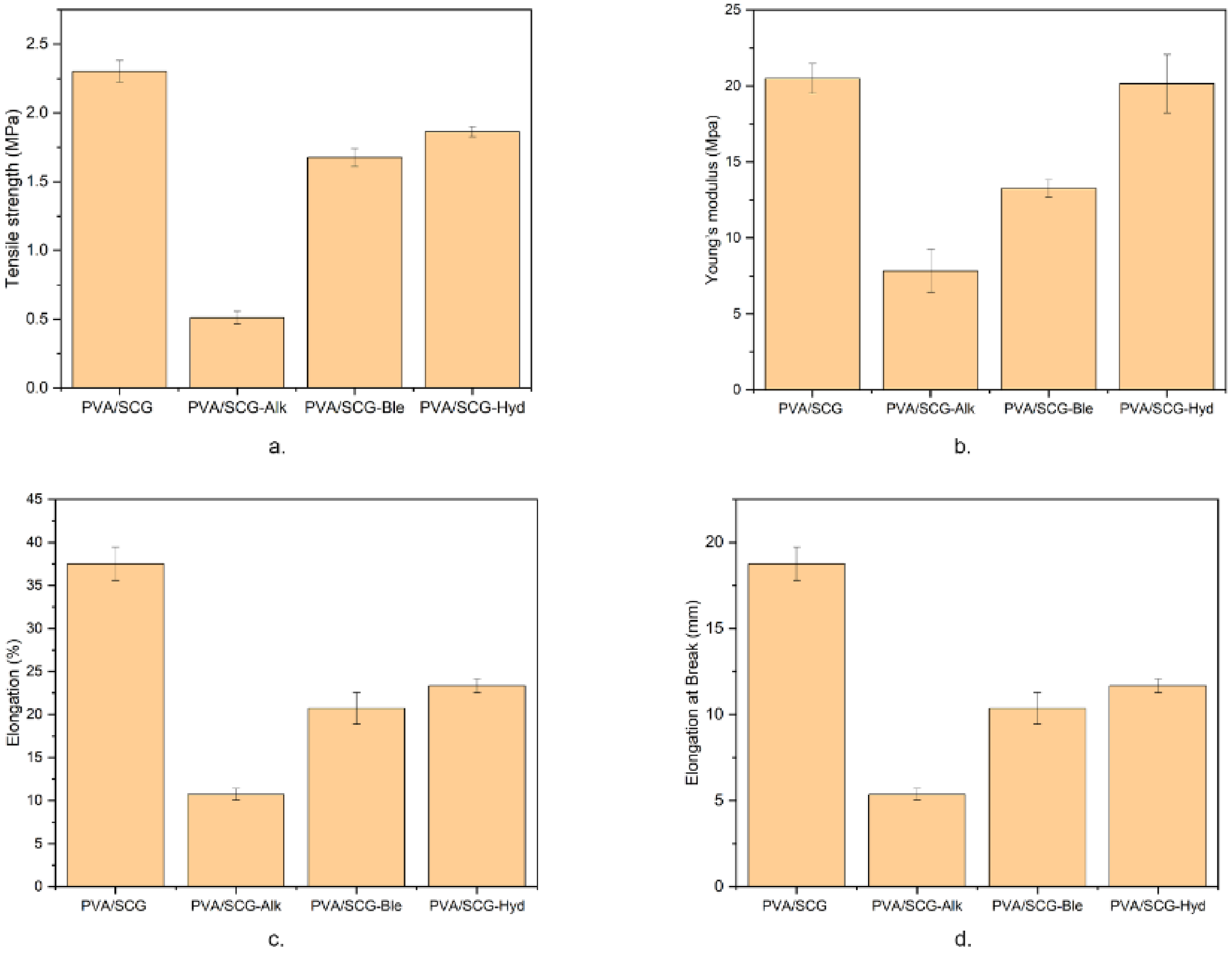

Fig. 7. Mechanical properties: (a) Tensile Strength, (b) Young's Modulus, (c) Elongation, and (d) Elongation at Break

In contrast, the PVA/SGC-Alk exhibited a drastic and significant deterioration in mechanical performance. Its maximum tensile strength plummeted to only 0.51 MPa, with a minimal elongation of 10.7%. This indicates that the alkalization treatment transformed the composite into a weak and brittle material. This dramatic decline is likely attributable to the degradation of the cellulose fiber structure under harsh Alkali conditions or the excessive removal of non-cellulosic components (such as hemicellulose and lignin), which weakened the filler's structural integrity and impaired its bonding with the PVA matrix [16], [24].

Meanwhile, the PVA/SGC-Hyd and PVA/SGC-Ble exhibited intermediate mechanical properties. The film produced from acid hydrolysis achieved a tensile strength of 1.86 MPa. This performance was slightly superior to that of the bleached PVA/SGC-Ble, which recorded a tensile strength of 1.68 MPa. Although both treatments yielded composites stronger than the alkali sample, neither could match the strength and the flexibility of the PVA/SGC sample. The minimal variation among samples within each treatment group indicates good consistency in the results.

Overall, these findings show that different chemical treatments of SCG impart varying effects on the composite's mechanical properties, confirming that not all modifications result in performance enhancement relative to the untreated control.

3.4 Tear resistance

The PVA/SGC exhibited the highest tear resistance with a value of 2.12 N (Table 1). This performance strongly correlates with the tensile test results, where this sample also showed the highest elongation. Ductile and flexible materials can dissipate energy at the tear tip through plastic deformation, requiring more energy to propagate the tear. The well-dispersed SGC likely also acted as crack bridges, effectively resisting further tearing.

Table 1. Tear resistance of PVA/SCG composite

Specimen name	Tear resistance (N)
PVA/SCG	2.12
PVA/SCG-Alk	0.78
PVA/SCG-Ble	1.66
PVA/SCG-Hyd	2.03

Conversely, the PVA/SGC-Alk sample displayed a significantly lower tear resistance of only 0.78 N. This drastic 60% decrease compared to the control confirms that the alkalization treatment rendered the material highly brittle. Brittle materials have a minimal capacity to deform and absorb energy, allowing tears to propagate easily with minimal force. This result is once again consistent with the brittle nature observed in the tensile tests.

The PVA/SGC-Hyd sample demonstrated excellent tear resistance at 2.03 N, nearly matching the PVA/SGC sample. This indicates that although the hydrolysis treatment slightly reduced the material's elongation, the resulting structure remained highly tough and effective at resisting tearing. On the other hand, the PVA/SGC-Ble sample had a lower tear resistance of 1.66 N. While still substantially better than the alkali sample, its value represents a noticeable decrease compared to both the hydrolyzed and control samples.

3.5 Discussion

3.5.1 Effect of alkali treatment

The mechanical tests revealed that treatment with 10% NaOH caused the most severe degradation in SCG composites. Compared to the PVA/SCG control, tensile strength, Young's modulus, elongation, and tear resistance decreased by 77.83%, 61.80%, 71.41%, and 63.21%, respectively (Fig. 6). The use of 10% NaOH in this study was based on previous literature, where this concentration has been widely applied to various lignocellulosic fibers such as banyan aerial roots, Napier fibre, and oil palm empty fruit bunches [9], [10], [11]. The present results demonstrate that the SCG was excessively harsh, leading to pronounced fiber degradation.

This finding is further supported by FTIR analysis. The disappearance of the aliphatic C–H stretching peaks at 2922 cm^{-1} and 2853 cm^{-1} after alkalization indicates an extensive removal of hemicellulose and lignin. These components typically reinforce the SCG structure and promote interfacial bonding with PVA. Their loss not only weakened the filler but also reduced its capacity for hydrogen bonding, thereby explaining the sharp decline in mechanical performance [23].

A study on Enset/Sisal fibers found that treatment with 10% NaOH caused severe structural damage, including fiber fracture and void formation due to fiber pullout, resulting in poor adhesion and a substantial decline in mechanical performance. By contrast, a milder treatment with 5% NaOH proved more effective, improving tensile strength by 5.2% compared to untreated composites [25]. Similar findings were reported for flax fibers, where exposure to 10% NaOH disrupted the straight fiber geometry, twisted the structure, and reduced the diameter by up to 51%, resulting in a drastic 58% loss in tensile strength and a 91% reduction in elongation at break [26]. Ikramullah et al. further emphasized that while a 5% NaOH treatment for 1 hour did not compromise the cellulose backbone, extending the duration to 2 hours risked damaging this key component essential for composite strength [24].

Consistent with these observations, other natural fibers also display fiber-specific responses to alkali treatment. For instance, screw pine fibers subjected to NaOH concentrations ranging from 2.5% to 15% underwent significant physical and chemical modifications; the highest concentration (15%) caused severe curling and structural collapse, while a moderate level of 5% drastically reduced lignin content (from 57.4% to 7.2%) and enhanced cellulose composition, thereby improving reinforcement potential [27]. In a related study, Benyahia et al. demonstrated that Alfa fibers treated with 7% NaOH for 24 h exhibited the best performance, with tensile and flexural strength rising by about 30% and 50% compared to untreated samples [28]. Conversely, work on bamboo fibers showed that although NaOH treatment at 1–5% improved interfacial adhesion and surface roughness, higher concentrations negatively affected yield strength and stiffness due to excessive lignin and hemicellulose removal [29]. These studies emphasize that the optimal concentration of NaOH is highly dependent on fiber type and morphology, and overly aggressive treatments tend to degrade critical structural components, ultimately weakening composite performance.

3.5.2 Effect of bleaching treatment

The SCG sample treated with sodium hypochlorite bleaching (PVA/SCG-Ble) showed reductions of 26.96% in tensile strength, 35.32% in Young's modulus, and 44.75% in maximum elongation. Although these decreases were less severe than those observed in the alkali-treated sample, they still indicate a significant decline in mechanical performance. FTIR analysis supports this result, as the disappearance of aliphatic C–H peaks after bleaching suggests that NaOCl not only removed impurities but also degraded hemicellulose and lignin. Such molecular damage weakened the fiber–matrix interface, thereby reducing the overall mechanical strength of the composites.

The literature indicates that high concentrations of NaOCl during bleaching can negatively affect the cellulose structure, the main component responsible for the mechanical strength of natural fibers. Aurelia et al. (2019) reported that treating fibers with 6% NaOCl for 3 hours at 60°C significantly decreased both cellulose content and yield. This was attributed to oxidative reactions that break glycosidic bonds in the amorphous regions of cellulose, thereby lowering its molecular weight and diminishing its ability to form hydrogen bonds with the PVA matrix. Consequently, the mechanical properties of the resulting composite films were adversely affected. The optimal treatment in their study was 10% NaOH without NaOCl (0%), yielding the highest cellulose content of 90.37%. In contrast, treatment with a combination of 10% NaOH and 6% NaOCl yielded only 66.58% cellulose content, highlighting the structural degradation caused by excessive bleaching [30].

Consistent results were also reported by Husnil *et al.* (2020) in their study on kenaf fibers. They found that bleaching with 10% NaOCl for 2 hours at 25°C reduced tensile strength due to cellulose depolymerization. High NaOCl concentrations led to oxidation of hydroxyl groups into carbonyl groups and cleavage of β-(1→4)-glycosidic bonds, ultimately reducing fiber diameter and structural integrity. In contrast, the most favorable condition was found at 1% NaOCl for 1 hour at 25°C, which effectively removed lignin and surface impurities without damaging the cellulose structure, thereby improving fiber strength and its suitability as a reinforcement material in composites [30].

Based on these findings, it can be concluded that optimal bleaching conditions are typically 1–3% NaOCl at 25 °C for 1–2 hours. Exceeding these parameters, particularly in terms of concentration (≥6%) or treatment duration and temperature, tends to result in irreversible cellulose degradation, ultimately impairing the mechanical performance of the final composite.

3.5.3 Effect of acid hydrolysis treatment

Acid hydrolysis treatment with 1 M H_2SO_4 on the SCG sample resulted in a composite with lower tensile strength (1.86 MPa) than the untreated control sample (2.30 MPa). FTIR analysis supports this result, as the acid-hydrolyzed SCG retained the C–H peaks at 2922 and 2853 cm^{-1}, suggesting that the treatment selectively targeted amorphous cellulose while causing minimal damage to lignin and hemicellulose. This preservation of aliphatic structures explains the relatively good tear resistance of the acid-treated composites and the absence of the extreme brittleness observed in alkali-treated ones.

Acid hydrolysis selectively targets and cleaves the β-1,4-glycosidic linkages in the amorphous regions of cellulose, while the more compact crystalline domains remain relatively resistant. This process induces two opposing effects. On one hand, the removal of amorphous regions increases the relative crystallinity index of the fibers, which may enhance stiffness and thermal resistance [31]. This could explain why the tear resistance of the hydrolyzed sample (2.03 N) remained high and was nearly comparable to that of the control (2.12 N).

On the other hand, acid hydrolysis primarily leads to depolymerization, reducing the length of cellulose polymer chains. Originally, long fibers, which can distribute stress effectively across the polymer matrix, are fragmented into much shorter particles known as microcrystalline cellulose (MCC). This significant reduction in fiber length, or aspect ratio (length-to-diameter ratio), adversely affects their reinforcing capability. Shorter fibers are less effective at transferring stress from the PVA matrix, thereby contributing to the overall reduction in tensile strength of the composite [32], [33].

Several studies have reported that incorporating MCC into PVA matrices does not always enhance mechanical strength. For instance, one study found that adding 5% cellulose to a starch/PVA composite decreased tensile strength from 8.6 to 5.2 MPa [34]. This decline is often attributed to MCC particle agglomeration within the matrix and poor interfacial adhesion. Unevenly dispersed MCC particles can act as stress concentration points rather than as reinforcing agents, ultimately triggering premature material failure [35].

4 Conclusions

The experimental results demonstrate that chemical treatments did not improve the mechanical performance of PVA/SCG composite films. The untreated sample exhibited the highest tensile strength and tear resistance, while all treated samples showed reduced properties, with alkalization being the most detrimental. FTIR analysis supported these findings, as the disappearance of the characteristic C–H peaks at 2922 and 2853 cm^{-1} after alkalization and bleaching confirmed the excessive removal of hemicellulose and lignin, indicating structural degradation of SCG. In contrast, acid hydrolysis preserved these peaks to a greater extent, resulting in less severe damage. Overall, these results demonstrate that the high concentrations of chemical agents used were overly aggressive, resulting in fiber degradation rather than enhancement. Future work should therefore focus on optimizing treatment conditions, particularly by reducing chemical concentrations, to preserve structural integrity and improve composite performance.

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