Characteristics of bulletproof plate made from silkworm cocoon waste: Hybrid silkworm cocoon waste-reinforced epoxy/UHMWPE composite

1 Introduction

In recent decades, the global incidence of terrorist attacks has surged, with Thailand among the nations witnessing irregular occurrences, particularly within its southern provinces. In response to this rising threat to human life, the deployment of protective armor has become imperative. Personal armor assumes a critical role in safeguarding individuals against projectiles, expertly countering penetration while mitigating impact forces. Historically, personal armor has been created from a range of materials including metal alloys, wood, glass, ceramics, steel, and aluminum [1]. Nevertheless, despite their efficiency in intercepting projectiles, these conventional materials are troubled by weight and thickness, thereby causing discomfort during prolonged wear and incurring significant costs.

In the search for optimal materials for personal armor, principal considerations encompass attributes such as lightweight construction, toughness, high tensile strength, and remarkable energy dissipation capabilities. Notably, recent research has shown increasing interest in natural fiber composites due to their potential to enhance ballistic impact [2–8]. The highlights are examples of natural fibers used as reinforcement in composites, such as jute, flax, and hemp, along with nonwoven flax and hemp combined with polypropylene serving as the matrix. Furthermore, it explores the use of jute and hemp fibers, sisal, bagasse, and alfalfa fiber in conjunction with epoxy resin. Distinguished by their lightweight structure, vigorous structural integrity, textured surface morphology, eco-friendly characteristics, and economic capability stemming from their utilization of recycled materials, these composites offer a compelling proposition. Moreover, they have commendable ballistic performance at a fraction of the weight and cost experienced by conventional counterparts [9,10].

Typically, an anti-ballistic plate comprises a multi-layered configuration. The initial layer, typically composed of either ceramic or natural fiber composite, functions to deform the tip of a ballistic projectile while effectively...
dissipating and absorbing their kinetic energy. Subsequently, a secondary layer, predominantly comprising materials such as Kevlar or ultra-high molecular weight polyethylene (UHMWPE), serves to further absorb residual kinetic energy. Researchers have extensively investigated various types of natural fiber composites integrated with aramid fabrics (i.e., Kevlar or UHMWPE) for ballistic impact applications [11–15]. Examples include composites reinforced with ramie fabric and epoxy, kenaf reinforced with virgin high-density polyethylene, curaua fiber reinforced with aramid fabric, coconut shell powder-epoxy composite reinforced with Twaron fabric, giant bamboo fibers reinforced with epoxy and aramid fabric, and sisal fibers reinforced with epoxy resin and aramid fabric. Although Kevlar/UHMWPE composites exhibit efficiency in impeding projectiles, their high cost remains a significant limit. In response, researchers are engaged in efforts to reduce this financial impact, whether through reducing the number of Kevlar/UHMWPE layers or substituting them with cost-effective natural fibers like silkworm cocoon waste. Silkworm cocoons, products of silkworm caterpillars, represent a unique natural structure filled with notable mechanical properties. However, the utilization of environmentally sustainable materials such as silkworm cocoon waste is being carried out with a focus on sustainability. Research conducted by Sashina and Yakovleva [16] underscores the significance of this attempt, revealing that silkworm waste constitutes a substantial proportion 50 wt% of the hy-products generated during silk production.

Silkworm cocoon waste, encompassing fragments, defects, discarded remnants, and unspooled husks following silkworm extraction, presents a viable option as a natural fiber residue. Its integration into epoxy resin composites not only satisfies the imperative for sustainable materials but also capitalizes on the abundant resources afforded by the silk industry. Moreover, silkworm cocoon waste exhibits distinctive attributes, including its lightweight nature and biodegradability, rendering it conducive to diverse applications. In the realm of composite fabrication, silkworm cocoon waste assumes a pivotal role as a reinforcing fiber due to its virtuous mechanical properties, notably, its toughness, which reduces the back face signature (BFS) of composite plates [17,18]. Epoxy resin, renowned for its exceptional adhesion, high strength, durability, and chemical resistance, serves as the matrix material. Nevertheless, the utilization of silkworm cocoon waste for ballistic applications remains relatively underexplored. Thus, the primary objective of this investigation is to fabricate a hybrid composite plate comprising UHMWPE/silkworm cocoon waste-reinforced epoxy composites via the cold pressing technique. This method, involving composite formation under ambient conditions using epoxy resins, is favored for its cost-effectiveness and simplicity. To assess the ballistic performance of the composite plate, a 9 mm, 8 g projectile was employed for impact testing. The fracture morphology of the natural fiber waste at the impact zone was investigated utilizing scanning electron microscopy (SEM). Subsequently, the ballistic efficiency of the hybrid silkworm cocoon waste-reinforced epoxy/UHMWPE composite (cocoon waste composite) specimens was compared with that of hybrid hemp woven-reinforced epoxy/UHMWPE composite (hemp woven composite), as hemp has garnered substantial attention and proven efficiency as a reinforcing constituent within polymer matrices in previous research activities [19,20].

2 Experimental

2.1 Materials

Bombyx mori silkworm cocoon waste was obtained from silk-producing communities located in the northeastern region of Thailand. These cocoons underwent careful preparation, involving the cutting of their tips and the removal of silkworm larvae from within. Concurrently, the hemp woven material was sourced from the hemp fiber product community enterprise situated in Chang Mai, Thailand. Both the silkworm cocoon waste and hemp woven specimens are depicted in Figure 1(a) and (b), respectively. The matrix employed in this study was epoxy resin YD 582, a modified bisphenol-A-based epoxy resin renowned for its vigorous properties. The hardening agent utilized, EPOTEC TH 7278, was a modified amine supplied by J.N. TRANSOS Company, Thailand. Additionally, the UHMWPE was sourced from a reputable Thai company based in Thailand.

2.2 Procedures

The compression molding technique was employed to fabricate the composite plate. In the preparatory phase, either silkworm cocoon waste or hemp woven material, weighing approximately 70 g, underwent compression via a hydraulic press at 6 MPa within a mold box measuring 15 cm in width, 15 cm in length, and 20 cm in height, resulting in the formation of a thin sheet. Particular care was taken to ensure uniform distribution of the silkworm cocoon waste, whereas the hemp woven material was cut to fit the dimensions of 15 cm × 15 cm. A hydraulic pressure of 6 MPa was applied to the mold for 1h. The UHMWPE was split into 15 cm × 15 cm sheets, totaling approximately 70 g in weight, as depicted in Figure 1(c).
Each sheet of UHMWPE was bonded together using epoxy resin. Subsequently, after complete adhesion of the UHMWPE sheets, either the silkworm cocoon waste or hemp woven thin sheet was positioned atop the UHMWPE layer. The epoxy resin was blended with the hardener in a weight ratio of 4:1, totaling approximately 240 g as a matrix solution. Before pouring, the matrix solution underwent degassing, followed by using a brush to eliminate air bubbles. The matrix solution was then poured onto the UHMWPE layer, along with either the silkworm cocoon waste or hemp woven sheet, within a mold box measuring 15 cm in width, 15 cm in length, and 5 cm in height, for 3 h. Subsequently, it was subjected to compression at 6 MPa for 12 h at ambient temperature. Upon removal from

Figure 1: Material for reinforced composite: (a) silkworm cocoon waste, (b) hemp woven, and (c) single UHMWPE sheet.

Figure 2: Composite plate samples before impact test with size 15 cm × 15 cm: (a) hybrid silkworm cocoon waste-reinforced epoxy/UHMWPE composite and (b) hybrid hemp woven-reinforced epoxy/UHMWPE composite.
the mold, the composite underwent curing at 80°C for 4 h. Finally, composite plate samples weighing a total of 380 g (i.e., cocoon waste composite) for ballistic protection with two layers were obtained, as depicted in Figure 2. The details and designation for compliant hybrid composites are defined in Table 1, and the graphical representation of these composites is represented in Figure 3.

### 2.3 Characterization of composite specimens

The tensile testing of samples was carried out at room temperature following the ISO-527-4 standard using a 30 kN system, which is an internationally recognized testing standard for assessing the tensile properties of fiber-reinforced plastic composites. Strength and modulus were assessed at a deformation rate of 2 mm/min. Each test included five samples, and the average values were recorded.

The composite plate samples underwent an impact test to assess the armor’s resistance to penetration. A 9 mm, 8 g weight ammunition was used for the test. The shooting device, comprising a gun barrel with a laser sight, was positioned 5 m away from the composite plate samples, as shown in Figure 4. The shooting was conducted horizontally perpendicular to the composite plate samples. The impact of kinetic energy absorption ($E_{\text{abs}}$) can be calculated using the following equation:

$$E_{\text{abs}} = \frac{m(v_i^2 - v_r^2)}{2},$$

where $m$ is the mass of the ammunition; $v_i$ is the projectile’s impact velocity; and $v_r$ is the residual velocity after the impact. The fracture mechanisms of the composite plate after the impact test were investigated by a JEOL JSM-S900LV SEM at an acceleration voltage of 20 kV.

The BFS, which is the extent of indentation in the clay witness caused by a nonperforating impact on the sample, was measured with a 0.01 mm Mitutoyo vernier caliper. The caliper wings were placed on the plane surface near the indentation rim. The depth caliper baseline was identified as this planar surface, as demonstrated in Figure 5.

### 3 Results and discussion

Hemp woven emerges as a natural material exhibiting considerable promise as a composite reinforcement agent, a conclusion substantiated by the investigations conducted by A. Consequently, in the context of this study, hemp woven was considered suitable for comparison with silkworm cocoon waste, given their shared natural origins. Tensile strength, denoting a material’s capacity to endure tensile forces along the vertical axis, and strain, representing its flexibility until reaching failure, constitute pivotal mechanical attributes. Examination of these mechanical properties, exactly outlined in Table 2, revealed noteworthy...
findings: the cocoon waste composite plate demonstrated a tensile strength of 107.4 MPa and an elastic modulus of 1240.3 MPa, slightly surpassing those of hemp woven. Our main focus in this study was to identify the material’s maximum tensile strength, which is a research goal corroborated by previous academic studies, especially those conducted by Han et al., Loh and Tan [18,21]. Although our presentation exclusively focuses on the tensile strength and elastic modulus values displayed in Table 2, with minimal discussion on the material’s behavior during testing, we acknowledge the necessity for further exploration in this field. Throughout the duration of this study, careful oversight was exercised in controlling the alignment, distribution, and volume fraction of reinforcement, aimed at optimizing tensile strength. As a consequence of our rigorous methodologies, the integration of cocoon waste reinforcement material yielded substantial enhancements in tensile strength, a phenomenon suitably substantiated by the scholarly observations made by Anidha et al. and Tanguy et al. [7,22]. Moreover, our previous investigation [23] corroborated the utility of silkworm cocoon waste as a reinforcement agent within an epoxy matrix. It demonstrated that this material contributes significantly to enhancing structural integrity. Specifically, the empirical evidence explained a remarkable increase of over 41% in tensile strength when the proportion of silkworm cocoon waste reached 42% of the composite weight, contrasting with the absence of such reinforcement.

The aim of this research is to develop simple and cost-effective composite plates. To achieve this, a method employing cold pressing was chosen to fabricate the composite plate

**Table 2: Mechanical properties of composite plate sample**

<table>
<thead>
<tr>
<th>Composite plate sample</th>
<th>Tensile strength (MPa)</th>
<th>Elastic of modulus (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hybrid silkworm cocoon waste-reinforced epoxy/UHMWPE composite</td>
<td>107.36 ± 2.03</td>
<td>1240.32 ± 1.38</td>
</tr>
<tr>
<td>Hybrid hemp woven-reinforced epoxy/UHMWPE composite</td>
<td>103.10 ± 1.38</td>
<td>1108.12 ± 2.59</td>
</tr>
</tbody>
</table>

**Figure 4:** Experimental setup of impact test.

**Figure 5:** BFS measurement.
material, enabling effective adhesion of the resin and reinforcement materials. Although the study did not investigate enhancing the adhesion properties between epoxy and UHMWPE, further exploration in this area was recommended. Two factors may contribute to the enhanced interfacial properties of cocoon waste composite. Chemically, cocoon waste fibers possess both hydrophobic and hydrophilic structural domains [24], facilitating polar-polar and hydrophobic interactions with the chemical groups in the epoxy matrix. However, morphologically, strong interfacial interactions may be hindered by an unfavorable contribution from the smooth surface morphology of silk fibers, as reported in previous studies [25]. It is hypothesized that in our scenario, hydrophobic interaction plays a crucial role, resulting in the observed improved interfacial characteristics of cocoon waste composite. The significant difference in hydrophobicity between epoxy and UHMWPE presents challenges in achieving strong bonding between these materials. While epoxy exhibits moderate hydrophobic properties, UHMWPE is highly hydrophobic, leading to limited affinity between their surfaces and hindering efficient adherence. Academic research has addressed this issue by exploring various surface modification techniques to enhance the compatibility of epoxy and UHMWPE. Plasma treatment, chemical functionalization, and the use of coupling agents have been investigated as methods to improve the bonding capacities of both materials by modifying their surface properties [26,27]. An alternative approach to this problem is to pretreat the UHMWPE, although this falls beyond the scope of this paper. Additionally, the tensile and elastic modulus properties of specific silk-reinforced composites described in recent literature vary depending on the matrix system employed. These composites utilized long continuous silk fiber [28], short-chopped fiber [29], and spun silk fabric [30,31] as the reinforcing phase. It is evident that the mechanical properties of these composites differ significantly based on the specific characteristics of the matrix system utilized. The adhesion between reinforcement and epoxy resin in composites involves various factors, including mechanical interlocking, chemical bonding, surface treatment, and surface morphology [32]. In the case of cocoon waste, a significant mechanism is mechanical interlocking, where the rough surface of the outermost sericin layer of the reinforcement material locks into the cured epoxy matrix, as evidenced by research findings from Morin and Alam [33]. While the composite tensile properties of silkworm cocoon waste may not exceed those of conventional epoxy resins, it offers advantages as a cost-effective and environmentally sustainable natural fiber. Moreover, when compared to hemp woven used as a reinforcing material, cocoon waste demonstrates superior tensile strength and elastic modulus.

Table 3 presents the results of impact velocity, residual velocity, energy absorption, and backface signature. Both composite plate samples demonstrate excellent projectile protection, with the bullet unable to penetrate the target. This suggests that the naturally reinforced composite effectively absorbs the projectile’s kinetic energy due to its remarkable hard and brittle properties, consistent with the findings of Hu et al. [34]. The energy absorption of the hemp woven composite slightly exceeds that of the cocoon waste composite, a trend supported by the result shown in Figure 6(b), indicating delamination and bending deformation, which contribute to the high absorbed kinetic energy of the projectile [2]. Additionally, residual kinetic energy is completely absorbed by UHMWPE. The BFS results of both composite plate samples are below 44 mm, meeting the standard set by the U.S. National Institute of Justice (NIJ) standard 0101.06 [35]. However, the BFS values of the cocoon waste composite and hemp woven composite differ slightly. The cocoon waste composite has a BFS of 21.25 mm, while the hemp woven composite has a BFS of 27.53 mm, as shown in Table 3. This composite plate consists of two layers: the first layer is natural fiber waste reinforced with epoxy resin, capable of absorbing significant energy and reducing the velocity of projectile impact, aligning with Drodge et al. [36]. The second layer is the UHMWPE layer, approximately 5 mm thick, which absorbs the residual energy of the projectile and provides a higher ballistic limit. Figure 6 illustrates the delamination phenomenon occurring in both composite plates, which is the predominant failure mode, initiated by the projectile’s compressive load at the impact point, leading to continuous stress that affects the debonding and fracture.
of UHMWPE fiber, consistent with the previous study by Suriani et al. [37]. The penetration process involves four steps. First, the projectile hits the composite plate, causing matrix cracking at the impact point. Second, the impact load from the bullet destroys the fiber bonding and causes breakage. Third, the composite absorbs the kinetic energy of the bullet, deforming its tip. Finally, the residual kinetic energy of the bullet is fully absorbed. The UHMWPE layer bulges and is not fully penetrated.

After the projectile attacked the composite plate samples, as illustrated in Figure 7, the results suggested that both composite plates effectively prevented further penetration. The projectile was stopped at the UHMWPE layer, as reported by Alkhatib et al. [38]. Figure 7(a) and (c) depicts full penetration without radial cracks, due to the inherently brittle epoxy resin and the toughness of both silkworm cocoon waste and hemp woven. Moreover, the robust interactions among silkworm cocoon waste, hemp woven, and the matrix contributed to a reduction in bullet velocity and substantial absorption of kinetic energy, resulting in relatively smooth damage at the impact point. However, the impact point hole of the cocoon waste composite exhibited more splaying compared to the hemp woven composite, attributed to the absence of weave in the silkworm cocoon waste, resulting in a rough and highly porous surface. In conclusion, both cocoon waste composite and hemp woven composite demonstrated effective performance as armor against projectile impacts. In Figure 7(b) and (d), bulges originating from the UHMWPE layer are visible in both composite plates, displaying pyramid-like deformation. Following the projectile’s penetration through the first layer, the UHMWPE layer fully absorbs the residual velocity. UHMWPE exhibits outstanding performance in both ballistic and projectile defense due to its high modulus and lower density [39].

The SEM image depicting the fracture of natural fibers at the impact point is shown in Figure 8. Fiber pull-out is observed in both composite plates. In Figure 8(a), the fracture pattern of the cocoon waste composite shows its rough surface, which encourages excellent adherence with the matrix and suggests the absence of bending deformation, thus enhancing the absorption of the projectile’s kinetic energy. Conversely, Figure 8(b) displays the smooth surface of hemp fiber due to its tightly woven structure, incorporated with hydrophilic properties resulting weak interactions with
the matrix. Bending deformation is visible in Figure 6(b). The distribution of both the matrix and cocoon waste fiber in the cocoon waste composites appears non-uniform across the epoxy resin surface, aligning with findings from Anidha et al. [7] and Yang et al. [40]. The interfacial connection between cocoon waste fiber and the epoxy resin matrix significantly influences the mechanical properties of the reinforced composite. Comprising fibroin fiber with a semicrystalline microstructure, the cocoon waste acts as a protein adhesive, enabling interfacial bonding with the matrix via hydrogen bonds [41]. This robust interfacial bonding greatly contributes to the strength of the closed-pack cocoon waste composite [42,43]. Consequently, both composite plate samples demonstrate effective performance in projectile protection, offering reduced cost, lightweight construction at 380 g per plate, and eco-friendliness through the utilization of cocoon waste and hemp woven from fabric production.

4 Conclusion

Composite plates were successfully manufactured using the cold pressing process, selected for their cost-effectiveness, simplicity, and adherence to sustainable manufacturing principles. Cocoon waste served a critical function...
as a reinforcement component in these composites. The fabrication process entailed mixing silkworm cocoon waste with epoxy resin and UHMWPE, followed by cold compression molding. Tensile and impact tests were subsequently conducted on the resulting samples, utilizing a standardized 9 mm projectile. The experiments yielded a tensile strength of 107.4 MPa and an elastic modulus of 1240.3 MPa.

The impact test results unequivocally highlight the remarkable projectile protection capabilities of both composite plate samples. The cocoon waste composite, leveraging the inherent strength and brittleness of its components, along with matrix interactions, effectively absorbed the kinetic energy of the projectile, echoing findings from previous studies. Conversely, the hemp woven composite, while demonstrating slightly superior energy absorption, also exhibited the interplay between delamination and bending deformation, leading to efficient kinetic energy dissipation. These results align closely with established standards for ballistic protection, as evidenced by the measured BFS values, all well below the U.S. NIJ standard 0101.06 of 44 mm. Despite their differences, the BFS values of the cocoon waste composite and hemp woven composite reflected their specific characteristics, with the former at 21.25 mm and the latter at 27.53 mm. Delamination emerged as a prominent failure mode in both composite plates, where continuous stress from the projectile’s compressive load triggers debonding and fracture of UHMWPE fibers. These findings collectively demonstrated that both the cocoon waste composite and the hemp woven composite offer promising and effective solutions for projectile protection. This research represents a crucial step toward the development of innovative, sustainable, and cost-effective materials for enhanced ballistic defense across various applications.

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**References**


