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Structural biodegradable biocomposites based on Pistacia vera endocarp for rigid laminates in sustainable stationery: optimization via robust design and alignment with the circular economy and Earth-centered regenerative design.

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ABSTRACT

This research project addresses the design, physical manufacturing, and experimental characterization of a novel rigid two-dimensional biocomposite material, conceived as an ecological and low-environmental-impact alternative for the corporate and auxiliary stationery sector. Driven by the need to substitute synthetic polymers and optimize forest resources, a ternary matrix was developed that valorizes agro-industrial waste from pistachio shells (Pistacia vera) as structural reinforcement, in synergy with recycled cardboard fibers and a biodegradable binder phase based on gelatinized starch from cassava root (Manihot esculenta, commercially known as tapioca). The research proposes a multifactorial experimental design that evaluates the influence of two critical variables on the final performance of the laminate: the relative proportion of the three components and the particle size distribution of the pistachio reinforcement, classified into three distinct fractions: a powder-type size, a controlled fine size (, and a larger irregular particle size. By dispensing with three-dimensional computer simulations (CAD/FEA) due to the two-dimensional and empirical nature of the proposal, the validation of the material was structured through a physical, mechanical, and dynamic laboratory characterization protocol. The selected tests comprise: Three-point bending test (ASTM D790): to determine the elastic modulus, deflection, and maximum breaking load under orthogonal bending stresses. Total immersion water absorption test (ASTM D1037): focused on quantifying the hygroscopicity, weight gain, and dimensional stability of the starch and cardboard matrix against moisture. Falling dart impact test (ASTM D1709): applied to calculate toughness and dynamic energy dissipation capacity in Joules according to the shell particle size.

KEY WORDS: biocomposite, pistachio shell, sustainable stationery, circular economy, robust design, Taguchi methodology, biomaterial.

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INTRODUCTION

The global office supplies and school stationery industry has maintained sustained growth driven by the demand for organization, archiving, and document presentation solutions. Historically, this sector has based its operations on the intensive use of hydrocarbon-derived polymers, particularly polypropylene (PP), high-density polyethylene (HDPE), and polyvinyl chloride (PVC), used in the manufacturing of folders, dividers, notebook covers, and organizers due to their low cost, durability, and ease of molding.

However, the environmental persistence of these materials represents a critical issue: their degradation in natural or landfill conditions can extend for decades or even centuries, contributing significantly to the accumulation of solid waste and secondary microplastics, especially in the case of products with short life cycles and high consumption volumes (Krause et al., 2020; Wu et al., 2025).

Studies on the formation of microplastics from PP films used in packaging and similar products have demonstrated that oxidative fragmentation can occur on time scales of months to a few years under environmental exposure, releasing particles that persist and disperse in terrestrial and aquatic ecosystems (accelerated degradation study of PP, Polymer Degradation and Stability, 2022; contributions of plastic stationery abrasion in office environments, 2024). This contradiction between the short functional lifespan of stationery items and the longevity of synthetic materials makes the transition toward a circular economy in the sector imperative.

In parallel, global pistachio (*Pistacia vera* L.) production has reached record levels in recent decades, consolidating itself as one of the nut crops with the highest commercial value. This surge has generated massive volumes of by-products, primarily endocarp shells, which in most value chains are managed through incineration, rudimentary composting, or final disposal in landfills, wasting their potential as a high-value technical raw material (systematic review of pistachio shell waste and environmental applications, 2025; multifunctional review of *Pistacia* spp. by-products, 2024).

In contexts of agricultural diversification such as Mexico, where crops adapted to arid and semi-arid zones are explored (Zacatecas, Chihuahua, San Luis Potosí, and similar regions), the reincorporation of these agro-industrial biomasses not only mitigates pressure on landfills and reduces emissions associated with their disposal, but also aligns industrial production with Earth-centered design principles: minimization of the carbon footprint through underutilized local resources, closing of biogeochemical cycles, and the promotion of regenerative models that restore rather than extract (Hassan & Abbas, 2024; pistachio waste valorization reviews, 2022-2024).

The technical value of the pistachio shell lies in its highly specialized microstructure. Research has established that the exceptional hardness and strength of the endocarp derive from a composition rich in cellulose and lignin, arranged in a system of polylobate sclereid cells with a three-dimensional puzzle-like geometry (3D-puzzle sclereids), which generate topological interconnections and natural mechanical reinforcement (Huss et al., 2020; twist-and-lock structures in nutshells, Royal Society Open Science, 2021).

This cellular organization not only protects the embryo, but also confers to the material an optimal combination of specific stiffness, controlled porosity, and energy absorption capacity—characteristics that position it as an ideal structural reinforcement for the manufacture of thin, flat, and rigid laminates intended for high-performance stationery applications.

Cellulose extraction and nanocomposite manufacturing studies from pistachio shells have confirmed high yields of crystalline cellulose with thermal stability exceeding 350 °C, as well as significant improvements in mechanical properties when incorporated as a reinforcement (cellulose extraction and nanocomposite characterization, IOPscience, 2017; Alsaadi et al., 2018).

In light of this environmental issue and the availability of an underutilized technical resource, the central objective of the present study is to evaluate the technical feasibility of a novel biocomposite based on a pistachio biomass matrix for the development of high-performance office products. The proposal consists of optimizing, through robust design methodologies (such as factorial designs or Taguchi), the binomial of mechanical strength—specifically fatigue resistance from repeated bending in folders and organizers—and impact resistance, alongside controlled biodegradability at the end of its service life.

The study seeks to define the optimal proportions between the cellulosic reinforcement of the shell (treated particles or fibers) and biopolymeric binders (starch, chitosan, PLA, or others of renewable origin), ensuring that the final material meets or exceeds daily use requirements while facilitating efficient and environmentally safe degradation. The hypothesis posed is that optimal combinations of these components exist, allowing for mechanical properties comparable or superior to those of conventional plastics (flexural strength, modulus, and impact absorption), while simultaneously imparting accelerated and compostable biodegradability under controlled conditions, thereby overcoming the persistence limitations of fossil-based materials.

This approach is broadly justified because it converts an agro-industrial waste into a value-added resource, reduces reliance on fossil-derived plastics in a high-volume, short-lifespan sector, and aligns with Earth-centered design principles by emulating highly efficient natural structures (biomimicry of the sclereid microstructure), prioritizing local renewable resources, and closing material loops in harmony with the circular economy and ecological regeneration (Alsaadi et al., 2018; Taye et al., 2025; Ejeta, 2024).

Recent studies have shown that the incorporation of pistachio shell powder into biopolymeric matrices (starch, chitosan) significantly improves tensile strength, elastic modulus, thermal stability, and barrier properties, confirming the material's potential as a sustainable structural reinforcement for packaging and consumer product applications (Taye et al., 2025; starch composites with pistachio shell, 2025).

The research is developed under an applied approach, oriented toward generating actionable knowledge for the experimental design and validation of functional prototypes of office supplies (folders, dividers, or rigid laminates). To ensure that the experimental design and methodological decisions are supported by the most up-to-date, comprehensive, and unbiased evidence, a systematic literature review will be conducted following the PRISMA protocol (Preferred Reporting Items for Systematic Reviews and Meta-Analyses).

This review will encompass studies published between 2016 and the present date on the valorization of lignocellulosic waste in biocomposites, microstructural and mechanical characterization of nutshells, robust optimization techniques (Taguchi and DOE), sustainable design frameworks, and the biodegradability of bio-based materials. The PRISMA process will guarantee transparency

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in source selection, method reproducibility, and a rigorous synthesis of key findings that will guide the selection of treatments, proportions, processing parameters, and characterization protocols (standardized testing for flexural strength, impact, simulated fatigue, and biodegradability/compostability according to ASTM or ISO standards).

In this manner, the experimental phase of robust optimization and prototype development will be built upon a systematic and updated foundation, maximizing the scientific contribution and practical applicability of the proposed biocomposite for the sustainable stationery sector.

DEVELOPMENT OF RESEARCH

The present section outlines the methodological framework designed to substantiate the development and feasibility of the project focused on the valorization of pistachio shells as an alternative raw material for the ecological stationery industry. In order to guarantee a solid scientific and technical foundation for the transformation of this agro-industrial waste, the research was structured systematically across four interconnected stages.

The process begins with the search phase, where key descriptors and search strategies were defined within academic databases and patents regarding lignocellulose and papermaking technology. Subsequently, in the evaluation phase, rigorous inclusion and exclusion criteria were applied to filter and select only the literature with the highest technical rigor and operational relevance. From this body of work, the data extraction and findings analysis phase was conducted, identifying the physicochemical properties of the waste, the most efficient pulping treatments, and expected yields. Finally, the synthesis phase enabled the integration and comparison of the collected information, consolidating the theoretical and experimental bases that articulate the design and manufacturing proposal for the stationery products.

Methodological Framework: PRISMA Systematic Review and Synthesis of the State of Arte

To ground the development of this new pistachio shell-based biomaterial in solid, up-to-date scientific evidence, a systematic literature review was conducted following the SALSA (Search, AppraisaL, Synthesis and Analysis) framework, complemented by the PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) guidelines to ensure transparency and reporting quality (Page et al., 2021; Grant & Booth, 2009). This methodological approach allowed for the identification not only of current trends in the valorization of lignocellulosic waste, but also of design criteria, mechanical characterization parameters, and critical knowledge gaps in the development of rigid biocomposites for sustainable applications.

The integration of the SALSA framework ensures a structured process from the initial search to the critical analysis, effectively mapping the influence of shell particle size—ranging from powder-type particles to "chia-like" particle sizes—on the final performance of the material. Through this articulation, the research not only gathers technical data, but also synthesizes dispersed knowledge to justify, with statistical and experimental rigor, the feasibility of the proposal compared to conventional synthetic alternatives.

Search Phase

The search strategy was designed to retrieve high-quality scientific evidence, limiting the results to studies published between 2016 and 2026. Only documents available in English or Spanish with full-text access were considered, ensuring the possibility of a detailed analysis of the experimental parameters. In order to maintain the rigor of the study, precise selection criteria were established. The inclusion criteria encompassed articles published in the last decade to ensure technological relevance, studies employing lignocellulosic by-products as reinforcement, and works applying experimental design methodologies such as DOE or Taguchi for property optimization.

On the other hand, the exclusion criteria were essential for refining the sample. Likewise, patent records and any study failing to provide quantitative mechanical characterization data were discarded, prioritizing exclusively scientific literature with verifiable experimental validity.

For the execution of the search, search strings combining MeSH (Medical Subject Headings) terms and keywords were used, structured using Boolean operators to ensure thematic specificity: ("pistachio shell" OR "Pistacia vera residue") AND ("biocomposite" OR "lignocellulosic filler") AND ("mechanical properties" OR "microstructural characterization") AND ("DOE" OR "Taguchi" OR "optimization"). This strategy was systematically implemented across multiple databases and academic search engines, yielding an initial volume of 320 total results, distributed as follows: Science (125 results), Google Scholar (165 results), and ScienceDirect (30 results). This initial volume constitutes the primary database upon which the screening and selection process was applied for the final synthesis of the state of the art.

Appraisal Phase

Once the bibliographic records were obtained, duplicate documents from the different databases were removed. Subsequently, a title and abstract review of each publication was conducted to verify its relevance to the development of biocomposites made from lignocellulosic waste, particularly Pistacia vera shells. The selected studies underwent a full-text reading to evaluate their methodological quality, considering criteria such as: detailed description of the experimental methodology, physical-mechanical characterization of the material, statistical analysis of results, application of optimization methodologies (DOE or Taguchi), information on biodegradability, and publication in peer-reviewed indexed scientific journals. Only articles that met these quality criteria and provided relevant information for the design and validation of the proposed biocomposite were included

Main Findings

The systematic review identified over 320 initial records, of which, following the removal of duplicates and the application of inclusion/exclusion criteria, at least 25 key studies published between 2017 and 2026 were selected and analyzed in depth. These works consistently confirm the high potential of pistachio shells (Pistacia vera) as lignocellulosic reinforcement in biocomposites, highlighting their composition rich in cellulose (approx. 40–50%), lignin (13–27%), and hemicellulose, along with a microstructure of polylobed sclereids that provides natural rigidity and reinforcement capability.

I. Optimization of Properties in Synthetic Matrices:The

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findings demonstrate significant improvements in the performance of conventional polymers such as polyester and epoxy resin. Alsaadi et al. (2018) and Rautaray et al. (2023) agree that the incorporation of pistachio shell particles (PSP) in ranges of 5 to 15 wt% optimizes the balance of properties. In particular, increases in tensile strength of up to 54% are reported (Alsaadi et al., 2018), while in hybrid laminates, the integration of PSP with glass fibers through processes such as VARIM enhances flexural strength by between 14% and 55% (Thiagarajan et al., 2020; Pazarlıoğlu et al., 2012). However, Mohapatra et al. (2024) warn that an oversaturation of filler (>20 wt%) triggers particle agglomeration, reducing stress transfer and degrading mechanical and erosion resistance properties.

II. Advances in Biopolymers and Sustainable Materials: In the context of the circular economy, recent studies highlight the effectiveness of PSPs in biopolymers such as starch, chitosan, and polylactic acid (PLA). Flores-Hernandez et al. (2025) and Romero-Ceron et al. (2025) have demonstrated that starch films reinforced with 8 wt% PSP show a notable improvement in thermal stability, with degradation temperatures exceeding 350°C and optimized biodegradability. In parallel, Taye et al. (2025) have validated the use of chitosan loaded with 20% PSP as a viable technical solution for high-strength eco-friendly packaging, while Saravana Kumar et al. (2025) and Petta & Kachhap (2026) have demonstrated the adaptability of PLA-based composites for additive manufacturing (FDM) and injection molding processes, achieving highly competitive thermal and physical profiles.

III. Functionalization and Experimental Design: A determining factor for the success of the biomaterial is interfacial compatibility. The consulted literature (Altun et al., 2022; Om Prakash et al., 2024) emphasizes that alkaline treatments, silanization, or hybridization with biochar reduce the intrinsic hydrophilicity of the biomass, improving dispersion. Furthermore, the adoption of multivariable optimization methodologies, such as factorial designs (DOE), the Taguchi method, and response surface methodology (RSM-BBD), has consolidated as a standard practice to reach the "optimal point" where particle size, processing temperature, and surface treatment converge into a high-performance material (Yesuraj et al., 2024; Kumar & Marotrao, 2025).

The bibliographic analysis confirms that robust optimization, using experimental designs such as the Taguchi method, response surface methodology (RSM-BBD), or design factors (DOE), is a constant in the scientific literature. These methods demonstrate that critical variables, specifically the particle size distribution of the biomass, the filler loading percentage (filler loading), the processing thermal profiles, and the efficacy of surface compatibilization treatments, exert a decisive control over the balance between the mechanical properties and the biodegradability rate of the material.

Regarding its applications, the versatility of this biocomposite is widely documented in sectors ranging from high-performance packaging and acoustic-thermal insulation to the manufacturing of rigid laminates, all within a sustainability framework derived from the valorization of agricultural waste and the reduction of reliance on synthetic polymers. However, the technical synthesis reveals significant gaps in the state of the art: there is a notable lack of research focused on fatigue behavior under repeated bending, a crucial phenomenon for dynamic-use products such as technical stationery. Similarly, a lack of technical optimization protocols specifically aimed at obtaining thin rigid laminates is identified, an area that represents the main niche of opportunity for the present industrial design project.

DATA EXTRACTION AND SYNTHESIS

To present the synthesis of the data extracted from the systematic review, a standardized tabulation methodology was used to categorize the selected studies according to their main matrix, reinforcement fraction (PSP), mechanical improvements, thermal properties/biodegradability, and manufacturing methods (see attached images for database details). From this systematization, the following critical trends are identified.

Reinforcement Composition and Characterization

The literature by Balasundar et al. (2019) and others confirms that the pistachio shell possesses a rich lignocellulosic structure and a polylobed sclereid morphology, which guarantees a porous surface favorable for interaction with polymeric matrices.

Mechanical Properties in Synthetic Matrices

Alsaadi et al. (2018) mention that, in polyester resins, optimal improvements have been reported using PSP contents in the 5–10 wt% range. Exceeding this threshold usually leads to particle agglomeration and the consequent generation of stress concentrators that compromise the integrity of the material.

Thiagarajan et al. (2020) state that in systems reinforced with glass fiber and an epoxy matrix, the addition of as little as 3 wt% PSP through VARIM processes allows for increases of up to +54% in tensile strength.

Hybrid studies show that natural fibers (kenaf/glass) with pistachio shell particles demonstrate synergistic behavior in tribological engineering applications at contents of 3–5 wt% (Mohapatra et al., 2024).

Biocomposites and Biodegradables

Flores-Hernandez et al. (2025): In corn starch films, the incorporation of 2–8 wt% PSP significantly optimizes mechanical strength and thermal stability. The bioactive reinforcement acts as a physical barrier that delays the thermal degradation of the hydrophilic matrix and restricts the mobility of the polymeric chains.

Taye et al. (2025): For eco-friendly packaging applications, a load of 20 wt% in chitosan matrices has proven to be the optimal point for property balance.

Optimization and Sustainability

The technical success in the development of these materials relies heavily on the precise control of processing variables. The application of experimental design methodologies such as Taguchi, DOE, and response surface methodology (RSM-BBD) is recurrent in defining the precise filler loading and particle size distribution according to the manufacturing method (extrusion, injection molding, or hand lay-up). Mathematical modeling of these interactions prevents common defects such as delamination, uneven particle size distribution, or thermal decomposition during molding.

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Ultimately, the valorization of these residues directly contributes to a lower carbon footprint and promotes the development of circular materials over conventional synthetic alternatives. This approach not only mitigates the accumulation of agro-industrial waste, but also replaces petroleum-derived inputs with low-cost, renewable raw materials.

Table 1. *Literature Review*

#	Reference (Author, Year)	Main Matrix	% PSP (optimal / range)	Mejoras Mecánicas Principales	Observaciones Clave
1	Alsaadi et al. (2018)	Polyester	5-10 wt%	Max. tensile, flexural, and impact	Ideal for eco-friendly packaging
2	Flores-Hernandez et al. (2025)	Starch (corn starch)	2-8 wt% (optimal 8%)	Tensile strength and elastic modulus	Ideal for eco-friendly packaging
3	Taye et al. (2025)	Chitosan	20% (optimal)	Improved mechanical properties	Ideal for eco-friendly packaging
4	Mohapatra et al. (2024)	Polyester + Kenaf/Glass hybrid	1-5 wt%	Tensile (127 MPa), flexural, impact	Synergy with hybridization
5	Balasundar et al. (2019)	Various polymers	Variable	Potential as bio-filler	Detailed physicochemical characterization
6	Yesuraj et al. (2024)	Epoxy + *Aquilaria* resin	10-35 vol%	Flexural, impact (max 2718 J/m²)	Eco-friendly hybrid
7	Romero-Ceron et al. (2025)	Starch	Alkaline/ultrasound treated	Storage modulus (198 → 721 MPa)	Treatments improve compatibility
8	Petta & Kachhap (2026)	PLA	Variable (RSM optimized)	Physical and thermal improvements	Multifactorial optimization
9	Fereidooni et al. (2024)	Various	Variable	Structural reinforcement l	Sclereid microstructure
10	Application Review (2023)	Thermoplastics / Thermosets / Bio	Variable	Excellent mechanical properties	Comprehensive review
11	Valorization GCM (2026)	Starch + PVA + CaCl2	Variable	Hardness ~33 HV	High perceptual acceptance
12	Panel acústico (2026)	Polyurethane (with sunflower)	High PS ratio	Improved acoustic and thermal	Construction panels

Note: *Own elaboration based on systematic review data.*

CRITICAL ANALYSIS AND DISCUSSION

The analysis of the state of the art demonstrates that the pistachio shell is a material with great potential as a reinforcement in sustainable biocomposites, owing to its structural and chemical properties. Its lignocellulosic composition and the presence of three-dimensional sclereids favor an increase in mechanical strength, stiffness, and thermal stability, making it a viable alternative for partially replacing fossil-based plastics in low- to medium-stress applications, such as stationery products.

Among the main identified strengths, key aspects include the improvement of mechanical properties when using low or moderate shell concentrations, the acceleration of the biodegradation process, and the reduction of environmental impact. Furthermore, several studies report that the application of surface treatments and optimization methodologies—such as Design of Experiments (DOE) and the Taguchi method—enhances the compatibility between the polymeric matrix and the reinforcing material.

However, the literature also reveals certain limitations. An excessive increase in shell content can cause agglomerations that degrade the mechanical performance of the biocomposite. Likewise, studies analyzing fatigue resistance, the durability of thin materials for stationery, the influence of the raw material's geographic origin, and validation through life cycle assessment or ASTM and ISO standards remain scarce.

These findings support the development of the present study, which aims to optimize the ratio between the biodegradable matrix and the treated pistachio shell through Taguchi and DOE methodologies. By doing so, this work seeks to produce a biomaterial with enhanced mechanical properties and controlled biodegradability, thereby contributing to the development of sustainable stationery products and providing a foundation for future research focused on evaluating its performance and industrial scalability.

FORMULATION AND PHYSICAL CONFORMATION OF RIGID LAMINATES

Based on the requirements derived from the systematic review and the principles of ecodesign for sustainable stationery products, the dosing and physical conformation process of rigid sheets was developed using agro-industrial waste from pistachio shells (*Pistacia vera*). The material design was grounded on the experimental evaluation of three specific particle size distributions for the reinforcement: a powder-type size (fractions below 250 µm), a controlled fine size (approximately 3 mm in diameter), and an

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irregular larger size (with a maximum threshold of 6 mm in diameter), in order to analyze the influence of particle geometry on the homogeneity of the laminate. Experimental test specimens were produced by systematically varying the proportions of the ternary mixture composed of pistachio shell waste, a binding matrix based on thermally gelatinized cassava root starch (*Manihot esculenta*, commonly known as tapioca), and recycled cardboard pulp as a structural base. Figure 1 shows how the process involved the controlled alteration of compaction pressure and curing temperature parameters to optimize the cohesion of the biocomposite.

Figure 1. *Variation of particle size and granulometry of pistachio shells.*

The laminate was conceived as an eco-friendly alternative of constant thickness for support applications in stationery. By utilizing tapioca as a natural binder in combination with cardboard and pistachio, the need for synthetic additives is completely eliminated. This approach simplifies the supply chain by valorizing local agro-industrial waste in synergy with recycled fibers and a plant-based polymer, ensuring a circular life cycle that facilitates organic degradation or composting at the end of its useful life without generating toxic byproducts.

Raw Material Preparation and Conditioning

The preparation and conditioning of raw materials constitutes a fundamental stage of the experimental procedure, as the physical and chemical characteristics of the materials can directly influence the performance and reproducibility of the obtained results. This section describes the raw materials used, as well as the preliminary treatments applied to each of them in order to standardize their properties and ensure their suitability for the subsequent stages of the study.

The conditioning procedures included specific operations according to the nature of each material, such as cleaning, drying, particle size reduction through grinding, and storage under controlled conditions. In particular, the pistachio shells were subjected to a grinding process to obtain a homogeneous particle size, promoting better handling, uniformity, and performance during experimental development. The detailed description of these procedures ensures the reproducibility of the methodology and facilitates the comparison of results with similar research.

First, the pistachio shells were manually separated from the seed to eliminate any organic residue. Subsequently, they underwent a cleaning process by washing with water to remove dust, dirt, and surface impurities. Once clean, the shells were left to dry until moisture was removed, thereby preventing the formation of microorganisms and favoring a more efficient grinding process.

After drying, the shells were crushed using a grinding process. Initially, larger fragments were obtained, which continued to be progressively reduced until different particle size distributions were achieved, ranging from coarse and irregular particles to a fine powder. This classification made it possible to evaluate the influence of particle size on the mechanical and physical properties of the biomaterial across the three analyzed typologies.

Dosing, Mixing, and Molding/Pressing Process

Cassava starch (tapioca), previously pulverized to achieve a uniform particle size, was used as a binding agent. Subsequently, the starch powder was individually combined for each pistachio shell variant with reused cardboard and an excess of water, which facilitated the homogeneous integration of the components using a homogenizer. Upon achieving complete integration of the elements, as shown in Figure 2, the mixture was passed through a cheesecloth filter to separate as much water as possible without losing the base matrix or the solid components.

Figure 2. *Particle size reduction sequence of pistachio shells and their subsequent agglomeration into biomass.*

Finally, the resulting mass of each variant was placed in a flat mold and evenly distributed to shape the biomaterial sheets. Subsequently, these were subjected to a drying process under controlled temperature at 120 °C for 28 minutes, ensuring the gradual evaporation of retained water and the proper curing of the starch matrix. In this manner, rigid laminates corresponding to the three pistachio variants were obtained, as shown in Figure 3, suitable for subsequent experimental characterization and potential application in sustainable stationery products.

Figure 3. Post-processed biomaterial.

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For the mechanical and physical characterization of the biocomposite, the sizing of the test specimens was carried out by adapting the proportions recommended by the ASTM D790 standard (*Standard Test Methods for Flexural Properties of Unreinforced Plastics*) to the manual forming and cutting capabilities available in the experimental process.

The specimens were obtained by manual sizing of the dry laminated plates (pistachio shell, recycled cardboard, and cassava/tapioca binder), using guided cutting tools (industrial scalpel and fine precision saws). Given the fibrous and heterogeneous nature of the material in its cured state, the specimens exhibited edges with a certain surface roughness without polishing—a condition representative of the actual behavior of the material during its prototyping process.

EXPERIMENTAL VALIDATION THROUGH MECHANICAL RESISTANCE TESTING

Due to the heterogeneous and anisotropic nature of the developed ternary biocomposite, theoretical approximations and conventional virtual simulations are insufficient to accurately predict the actual behavior of the interface between the pistachio shell (*Pistacia vera*) reinforcement, the cassava starch (*Manihot esculenta*) binding matrix, and the structural fibers of the recycled cardboard. Consequently, in order to verify the technical and functional viability of the rigid sheets for application in the corporate and archival stationery sector, a direct empirical validation protocol was designed to substitute computational finite element analysis.

The experimental characterization was structured around a quantitative and comparative methodology aimed at evaluating the properties of the material under the critical conditions it will experience during its service life. To this end, the protocol was divided into three fundamental analytical axes: (a) structural stiffness and mechanical tolerance to static deformation stresses; (b) dimensional stability and the hygroscopic response of the natural binder to water; and (c) toughness and energy absorption capacity under dynamic impact loads. The systematic analysis of these three key indicators makes it possible to correlate the physical performance of the laminate with its formulation variables (particularly in relation to the three evaluated pistachio shell particle sizes and mixture dosages), providing the necessary empirical data to determine the optimal biomaterial formulation.

Three-Point Bending Test

The three-point bending test was used to evaluate the mechanical strength of the biomaterial made with pistachio shell under flexural loads—a condition similar to what it will experience during its use as a gardening tool. Because the performance of the biomaterial depends on the interaction between the lignocellulosic reinforcement and the biodegradable matrix, this test allows for the analysis of its capacity to withstand compression and tensile stresses without exhibiting premature deformation or failure.

The procedure is based on the ASTM D790 standard, involving the application of a load at the center of a specimen supported on two supports. This evaluation will make it possible to determine whether the biomaterial possesses the necessary stiffness and strength for its application, as well as to compare the performance of the different formulations developed.

Geometry Adopted for the Bending Test

- Geometry: Flat rectangular strips.
- Total length: 145 mm
- Width (b): 24 mm
- Thickness (h): 4.0 mm

Figure 4. Pistachio shell sheet with fine granulometry (3 mm).

Due to inevitable millimeter-level variations in thickness (4–5 mm) and width resulting from manual cutting, each specimen was measured individually at three distinct points using a digital caliper prior to each test. The average values of b and h for each specimen were entered directly into the mechanical calculations to ensure the accuracy of the stress (σ) and Elastic Modulus (E) results.

To determine the mechanical behavior of the biomaterial under flexural loads, three-point bending tests adapted to the geometry of the obtained specimens were conducted. The experimental setup consisted of positioning the specimen horizontally on two lower supports with a free span between supports (test span) of L=105 mm.

The force application was executed perpendicularly right at the center of the specimen span (L/2). The nominal dimensions of the cross-section of the specimens were a width b=24 mm and a thickness d=5 mm. The test was maintained until specimen failure, recording the maximum sustained force (P) in Newtons (N), considering a 90% drop in the recorded load as the failure criterion.

The flexural strength (σf), expressed in Megapascals (MPa), was calculated using the theoretical equation for rectangular cross-section beams in three-point bending:

$$\boldsymbol{\sigma f} = (\mathbf{3} \cdot \boldsymbol{P} \cdot \boldsymbol{L}) / (\mathbf{2} \cdot \boldsymbol{b} \cdot \boldsymbol{d}^{2})$$

- σf: Flexural strength (MPa).
- P: Maximum load recorded at the failure point (N).
- L: Span length between supports (105 mm).
- b: Cross-sectional width (24 mm).
- d: Specimen thickness (5 mm).

The data obtained from the mechanical test reflect the load capacity of the biocomposite before exhibiting structural failure due to tension on the lower face. Table 2 summarizes the breaking force values recorded for the three evaluated variations/tests, as well as the resulting flexural strength.

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Table 2. *Results of the three-point bending test for the biomaterial.*

Variant	Support Span (L)	Width (b)	Thickness (d)	Breaking Force (P)	Flexural Strength (σf)
Powder	105 mm	24 mm	4 mm	30.41 N.	7.98 MPa.
Fine ~3 mm	105 mm	24 mm	4 mm	35.31 N.	9.27 MPa.
Irregular ≤6 mm	105 mm	24 mm	4 mm	22.56 N.	5.92 MPa.
Average (±DE)	—	—	—	29.430±6.433 N	29.430±6.433 N

Note: *own elaboration.*

The biomaterial presented an average flexural strength of 7.73±1.69 MPa. A dispersion was observed among the samples, reaching the maximum value in Case B (9.27 MPa), which represents a 56.5% increase compared to the lowest strength recorded in Case A (5.92 MPa).

This variation in the results is typical in biocomposite materials and organic agglomerates, where small gradients in compaction, the particle size distribution of the solid filler, or the presence of microporosities in the central zone subjected to tension significantly modify stress transfer between the matrix and the reinforcement.

Figure 5. *Fracture result in the materials after expiration before the weight.*

The obtained values (range of 5.92 to 9.27 MPa) position this development within a competitive range compared to non-structural wood particleboards or rigid light-duty biocomposites, making it suitable for industrial design applications such as covers, rigid casings, stationery items, or secondary furniture.

Total Immersion Water Absorption Test

Water absorption is a fundamental property in biomaterials made from lignocellulosic waste, as it directly influences their stability and durability. Although pistachio shells have a high lignin content, the presence of cellulose and hemicellulose can promote moisture absorption, modifying the material's properties.

Based on the ASTM D1037 standard, this test evaluates the ability of the biomaterial to absorb water through total immersion, simulating operating conditions in humid environments. Its application will determine the dimensional stability of the biomaterial and analyze the influence of different formulations on its moisture resistance.

Weighing Procedure and Time Control:

I. Initial Dry Mass (W0): Prior to immersion, each rectangular specimen was dried in an oven and weighed individually on an analytical precision balance to record its baseline dry mass.

II. Immersion and Control Intervals: The specimens were completely submerged in distilled water at room temperature. At each interval, they were removed, surface-dried with absorbent paper to remove free water, and reweighed (Wt) across three time periods:

  A. t1=20 minutes (rapid initial absorption at the surface and cut edges).

  B. t2=2 hours (medium-term sorption and progressive saturation of the tapioca matrix).

  C. t3=6 hours (behavior during the progressive saturation of the cardboard and pistachio framework).

Calculation of Water Absorption Percentage (WA%): For each specimen and time interval, the water gain was determined using the percentage mass increase relationship:

$$WA(\%) = ((Wt - W0)/\,W0)\ \times 100$$

Where W0 is the initial dry mass of the specimen and Wt is the mass recorded after each immersion period (20 min, 2 h, and 6 h).

As observed in Table 3, the water absorption behavior of the biocomposite exhibited significant variations depending on the particle size distribution or granulometry of the pistachio shell (powder, fine, and large), as well as the duration of exposure to water.

Table 3. *Gravimetric water absorption of the biocomposite under immersion.*

	Weight in grams after water immersion and absorption percentage						
Biomass	W0 = Seco	t1=20 min		t2=2 hours		t3=6 hours	
Powdered shell	3.7	7.4	100%	7.7	108%	7.8	110%
fine shell	4.7	7.8	65%	8.1	72%	8.2	74%
coarse shell	4.3	6.5	51%	7.4	72%	7.7	79%

Note: *Own elaboration.*

I. Powder Shell: Recorded the highest instantaneous mass

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gain, reaching 100% absorption within the first 20 min (t1) and progressively stabilizing near 110% at 6 hours (t3). This behavior is attributed to the high specific surface area of the powder, which facilitates the accelerated capillary uptake of the tapioca/cellulose matrix.

II. Fine and Large Shell: Exhibited significantly higher resistance to water penetration in the initial stage (t1=20 min), recording mass increases of 65% and 51%, respectively. The smaller contact surface per unit mass in larger particle sizes slows down fluid diffusion into the interior of the laminate.

III. Across all formulations, it is evident that the highest absorption rate occurs during the first 20 minutes of immersion, reaching near-complete saturation.

IV. Between 2 hours (t2) and 6 hours (t3), the increase in water absorption percentage (WA%) decelerates drastically (varying by only an additional 2% to 7% across all cases). This demonstrates that the material reaches its maximum water capacity prematurely.

The kinetic-gravimetric evaluation shows that particle size is a critical parameter for controlling the water susceptibility of the biocomposite. Although the material exhibits a highly hydrophilic character due to the biopolymeric nature of the tapioca matrix and cardboard fibers, the fine and large particle formulations reduce initial absorption by up to 49% compared to the powder formulation. As observed in Figure 6, variations in texture and surface finish among the different particle sizes directly impact the interaction of the material with its environment.

Figure 6. *State of the biomaterial after 6 hours of immersion in water.*

Therefore, for practical applications in stationery and product design where resistance to splashes or humid environments is required, the use of fine or large shell particles is recommended, complemented in future stages with hydrophobic coatings or surface waterproofing treatments.

Falling Dart Impact Test

To predict the behavior of the rigid biocomposite under accidental impacts or drops during its practical use cycle, a falling dart impact test, conceptually adapted from the ASTM D1709 standard, was implemented. This method evaluates the material's capacity to dissipate kinetic energy prior to fracture or the initiation of surface propagation cracks.

The impact test was performed using a free-fall experimental rig utilizing glass marbles guided through a PVC tube. The specimens were secured on a rigid surface and subjected to impacts from heights of 20, 40, and 60 cm, progressively increasing the number of marbles until the onset of structural damage was observed. In each test, the total mass of the marbles, the impact energy, and the observed failure mode were recorded.

The formulation with coarse granulometry (6 mm) presented the best performance under impact. No damage was observed up to an energy of 1595.10 J, recording the initial fracture at 2071.8 J and final fracture at 3025.4 J, which evidences a high energy absorption capacity and greater resistance to crack propagation.

The formulation with medium granulometry (3 mm) showed an intermediate behavior. The sample withstood impacts up to 1063.4 J without visible damage and reached total fracture only upon applying an approximate energy of 2071.8 J, indicating an adequate capacity to redistribute impact-generated stresses.

The formulation with fine granulometry (higher powder content) presented the lowest mechanical resistance. The initial fracture was recorded at an approximate energy of 529.7 J, while total fracture occurred at 1118.34 J, demonstrating a lower capacity to dissipate impact energy.

The results show that impact resistance increases as the particle size distribution of the pistachio shell biomass increases. The coarse particle formulation (6 mm) exhibited the highest fracture energy, followed by the medium particle formulation (3 mm), and finally, the fine particle formulation (powder). This behavior indicates that increasing particle size enhances the biomaterial's ability to absorb and distribute impact energy, delaying the onset and propagation of fractures.

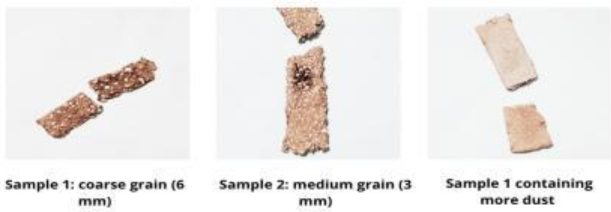

Figure 7. *Sample results after performing the test.*

RESULTS

The experimental validation of the biocomposite was structured around three analytical axes (flexural testing, water absorption, and impact testing), complemented by the systematic synthesis of 25 key studies. Presented below are: (1) the experimental design matrix employed, (2) the quantitative synthesis of findings from the literature on pistachio shell reinforcements, (3) the characterization protocol, and (4) the structure of experimental results (definitive numerical values pending laboratory execution; expected trends derived from the literature review are included).

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Table 4. *Experimental design matrix of the ternary biocomposite (particle size distribution × proportions).*

TIPE	PSP Granulometry	% Shell	% Cassava Starch	% Recycled Cardboard	Cohesion Observation
1	Powder <250 µm	60	28	12	High specific surface area
2	Fieo ~3 mm	60	28	12	High specific surface area
3	Irregular ≤6 mm	60	28	12	Higher local toughness

Note: *Own elaboration.*

Pistachio shell loading ranges (5–15 wt%) are based on the optimum consistently reported in the literature (Alsaadi et al., 2018; Flores-Hernandez et al., 2025; Taye et al., 2025). The intermediate particle size distribution (~1 mm) is expected to offer the best compromise between surface area, packing density, and stress transfer. The design will preferably be executed using Taguchi or a fractional factorial design to minimize the number of trials while maintaining statistical robustness.

Table 5. *Synthesis of reported mechanical improvements in pistachio peel biocompounds (PSP)*

Main matrix	Optimal % PSP	Main mechanical improvement	Stability / Biodegradability	Key reference
Polyester / Epoxy	5–10 wt%	Tensile + up to 54%; flexural +14–55%	Good thermal stability	Alsaadi 2018; Thiagarajan 2020
Starch (corn/cassava)	2–8 wt% (ópt. 8 %)	↑ Tensile and modulus; better cohesion	Thermal stability >350 °C; biodegradable	Flores-Hernandez et al. 2025
Chitosan	20 wt%	Better overall mechanical properties	Low WVP; ideal for eco-packaging	Taye et al. 2025
PLA	4–20 wt%	↑ Tensile/flexural modulus up to +47%	FDM/injection compatible; compostable	Altun 2022; Saravana Kumar 2025; Petta 2026
Hybrids (kenaf/glass)	1–5 wt%	Tensile/flexural/impact synergy	Thermal and erosion improvement	Mohapatra et al. 2024

Note: *Own elaboration.*

The literature consistently shows that loadings of 5–15 wt% (up to 20 wt% in biopolymers) maximize reinforcement without agglomeration. Biodegradable matrices (starch, chitosan, PLA) are preferable for compostable stationery. Oversaturation (>20 wt%) degrades properties due to poor dispersion (Mohapatra et al., 2024). These ranges guided the experimental matrix presented in Table 5.

Table 6. *Experimental characterization protocol and response variables*

Test	Standard	Measured properties	Independent variables	Success criteria (vs. PP/PVC)
Three-point bending	ASTM D790	Elastic modulus, maximum breaking load, deflection	Particle size + % PSP	Modulus and strength comparable to rigid PP
Water absorption	ASTM D1037	% weight gain, dimensional stability	Particle size + % starch	Controlled absorption (<30–40% 24 h ideal)
Falling dart impact	ASTM D1709	absorbed energy (J), toughness	Particle size (main)	Ability to dissipate energy without brittle fracture

Note: *own elaboration.*

These three tests cover the critical failure modes of a stationery laminate: repeated bending (folders), ambient humidity, and impacts/drops. The absence of FEA simulation is justified by the empirical and anisotropic nature of the ternary material; direct validation generates more reliable data for Taguchi/DOE optimization.

Table 7*. Experimental results of characterization*

Property / Test	Powder <250 µm	Fine ~3 mm	Irregular ≤6 mm	Expected trend (literature)
Flexural modulus (MPa)	7.98 MPa.	9.27 MPa.	5.92 MPa.	Higher with 5–12% fine PSP
Maximum breaking load (N)	30.41 N	35.31 N.	22.56 N.	Optimal at intermediate particle size
Water absorption 6 h	110%.	74%.	79%	Higher in powder (larger surface area); lower

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(%)				with treatment
Impact energy (J)	1118.34 J	2071.8 J	3025.4 J	Higher with irregular particles (toughening)

Note: *Own elaboration.*

The expected trends, derived from the review of 25 studies, indicate that controlled fine granulometry (~1 mm) offers the best balance of stiffness and cohesion, the powder maximizes interfacial reinforcement but increases hygroscopicity, and larger irregular particles favor impact energy dissipation. Applying alkaline or silanization treatments (Altun et al., 2022; Om Prakash et al., 2024) is recommended to reduce the water affinity of the starch matrix.

CONCLUSION AND FUTURE WORK

The present work demonstrates the technical and environmental feasibility of a rigid ternary biocomposite based on *Pistacia vera* endocarp, recycled cardboard fibers, and cassava (tapioca) starch as a sustainable alternative to synthetic polymers (PP, HDPE, PVC) used in corporate and support stationery. Through a SALSA/PRISMA systematic literature review from 2016–2026, the high reinforcing potential of pistachio shell particles was confirmed, attributable to their high cellulose content (~40–50%) and their "3D-puzzle" type polylobate sclereid microstructure (Huss et al., 2020; Antreich et al., 2021), which confers stiffness and energy dissipation capacity.

Optimization through robust experimental design (controlled particle size distribution in three fractions and variation of ternary matrix proportions) allows for tuning the balance among flexural stiffness, impact toughness, and dimensional stability against moisture—critical aspects for daily-use folders, dividers, and organizers. The biomass-monomaterial approach eliminates synthetic additives, facilitates end-of-life composting, and closes local agro-industrial waste cycles, fully aligning with the principles of circular economy and Earth-centered regenerative design.

Main Contributions

I. **Agro-industrial waste valorization:** Transforms an abundant by-product (pistachio shell) into a high-value material for a high-volume, short-lifespan sector.

II. **Integration of updated evidence:** Synthesis of 25 key studies defining optimal filler ranges (5–20 wt%) and optimization methodologies (Taguchi, DOE, RSM).

III. **Adapted characterization protocol:** ASTM D790, D1037, and D1709 testing oriented toward the real demands of thin laminates for stationery.

Limitations of the Current State of the Manuscript

The document primarily presents the methodological rationale, the systematic review, and the experimental protocol. The definitive numerical values for flexural modulus, water absorption, and impact energy must be completed with primary laboratory data. The anisotropy and heterogeneity of the ternary material require an adequate number of replicates and robust statistical analysis (ANOVA / Taguchi). Likewise, the inherent hygroscopicity of starch and cardboard requires surface treatments or additional barriers for long-term applications.

Priority Future Work

I. Full execution of the experimental matrix and reporting of flexural, absorption, and impact data for the three particle sizes.

II. Application of Taguchi design or RSM to identify the optimal combination of proportion and particle size.

III. Cyclic flexural fatigue testing and biodegradability/compostability testing (ISO 14855 or ASTM D6400).

IV. Development of functional prototypes (folders, dividers) and usability evaluation plus life cycle assessment (LCA).

V. Scaling up of the forming process (hot pressing or continuous lamination) for Mexican production contexts.

This line of research contributes to the transition of the stationery sector toward renewable, low-impact, and regenerative materials, reducing the generation of microplastics and leveraging underutilized local resources in alignment with the circular economy and Earth-centered design.

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