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Sustainable innovation in Pulp and Paper industry: Invasive alien species *Phytolacca americana* waste integration to commercial pulp. Physic-mechanical properties analysis

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Abstract

Paper industry contributes significantly to the transformation of native forests into monocultures of exotic species, such as *Eucalyptus globulus*. This monoculture expansion not only reduces biodiversity in cultivated areas but also in adjacent regions. Ecological concerns, the demand for cellulose-based products continues to rise, driving an increase in pulp and paper production over the years.

Invasive species pose a significant threat to ecosystems, driven by anthropogenic behaviors, urbanization, cultivation, and globalization. The integration of these species leads to the displacement of native flora, soil impoverishment, and ecosystem degradation.

Addressing both issues highlights the need not only to reuse waste derived from clearing invasive species but also to reintroduce it into the production system by utilizing their fibers reducing the use of alien wood fibers in paper production. The study encompasses the entire process of obtaining and processing *Phytolacca americana* pulp, including kraft lignin extraction, refining processes, and analysis of pulp and laboratory paper properties. Standardized tests based on ISO standards were conducted to assess various physical and mechanical properties of the paper samples such as drainability, thickness, grammage, specific volume and density, weight, and air permeance and resistance. Advanced imaging techniques as SEM visualization were employed to examine the internal structure and chemical composition of the paper samples, providing valuable insights into fiber integration and paper quality.

Results confirm the suitability of *Phytolacca americana* utilization for industry, with certain properties showing improvement or maintenance, indicating its feasibility for market incorporation, promoting biodiversity care and monoculture reduction.

Keywords: Invasive Alien Species; paper production; biomass reutilization; non-woody pulp integration;

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1. Introduction

Paper and pulping industries are extremely related with environmental and ecosystems health. Paper industry is not only the fifth sector most pollutant, generating an amount of 220-380 m³ of highly coloured and potentially toxic wastewater (Badar & Haq 2011); but it is responsible of most monocultures production, consuming the 40% of world's commercially cut timber. Apart from this, paper production remains one of the most relevant industries globally, acquiring a total amount of 2.519.089 € in past 2018 only in Spain (MITECO) and as stated by the As Papel Association, in 2022, Spain's overall production reached 6.3 million tons, with 1.5 million tons designated for pulp and 6.3 million tons for paper and cardboard. This situation positioned Spain as sixth in paper and fifth in pulp production within the EU. According to preliminary CEPI reports in 2021, paper industry has increased in a 5.8% in relation with previous year (Vrabic-Brodnjak & Mozina 2022).

Those wicked situations contribute to the deterioration of natural ecosystems surrounding factory mills and pulpwood plantations. Paper mills are usually located near wood plantations in order to decrease costs; natural forests were usually used for papermaking industry in the past, but, now a days, the use of natural forest is being substituted by plantations of fast-growing growing species, usually exotic, such as Eucalyptus and Pines. Planted forest were, in 2014, the 7% of total global forest area, or 264 million ha. (Area 2014).

According to the Annual Forest Statistics of 2018, an amount of 33% of timber is destined to pulp production, being the highest proportion of wood commercial uses. In comparison, in the case of sector, the 38% of wood is destined to bioenergy, in front of 21% for pulp production (MITECO).

At the north of Spain, specifically in Galicia, there is a volume of timber areas of 125m³/ha in front of 189 m³/ha of forest surface. Compared with this, south of Spain present an average of 20 m³/ha – 50 m³/ha (MITECO). A third part of total forest plantation in Galicia is destined to commercial deforestation. The three principal arboreal species in Spain are from Pinus family, but fourth one, even exotic, is Eucalyptus globulus occupying 79.645.351 m³ c. c. in all Spanish surface (MITECO), part of the forest area destined to production, a 8% of total forest area, specifically 619.918 ha.

Eucalyptus plantations proliferation in Galicia are derived from ENCE, Pontevedra's paper mill, which in 2022 spent 236.000.000€ in timber from individuals (Merino & Gil 2023), covering the 95% of demand needs. Other studies demonstrated that a third part of Galician Forest has been planted by humans, with commercial ends.

Eucalyptus species demonstrated not only an expansive behavior but invasive, preventing local species to grow, due its leaves chemical composition, which intervenes in soil composition, increasing its pH and avoiding biodiverse flora proliferation (Falkiner & Smith 1997). Even now a days, several studies (Álvarez et al 2021; Iglesias et al 2023) demonstrated high possibilities for Eucalyptus industry waste reutilization for antioxidant extraction and papermaking, main problem remains in high commercial demands that are translated into monocultures incrementation (Merino & Gil 2023).

Furthermore, monocultures problematic in relation with biodiversity flora, forest urbanization and anthropogenic factors as agriculture and land management's result in ecosystem diversity loss, favoring exotic and invasive species proliferation (Valero et al 2015). Local vegetation in riparian areas is being substituted by invasive species such as selected for this study *Phytolacca americana*. (Álvarez 2015). The impact of Invasive alien species in natural ecosystems, disturbing its balance through nutrients and resources competition, diseases transmission, intertwining in soil and light conditions, provoking the reconfiguration of the ecosystem functioning (Vrabic-Brodnjak & Mozina 2022; Ordóñez & Duinker 2012)). Major presence of Invasives Plant species in managed environments tends to have consequences in the ecological integrity of the ecosystem, altering the natural composition of species and habitat (Conway et al 2019; Goodwin & Shriver 2014).

According to the European Commission, there are 41 plant species that suppose a threaten for European natural ecosystems biodiversity. In Regulation (EU) 1143/2014, Invasive alien species are presented as one of the main threats to biodiversity and related to ecosystem services, which are being intensified by global trade, transport, tourism, and climate change, consisting of a cross-border challenge affecting the whole Union (European Commission 2014). When referring to invasive alien species, it is relevant to confirm anthropogenic introduction and naturalization in different ecosystems than its original habitat; by human means, this species were usually introduced for cultivation or for ornamental means and derived into ecosystem degradation (European Commission 2014). In affected ecosystems, rapid restoration and

mitigation of Invasive species is crucial for flora maintenance, for this reason, several management activities are being performed all around affected areas, such as the one selected for this study, in Ribadumia, Galicia. Derived from this management initiatives, promoted by city council and governmental decisions, amounts of fiber waste are being generated, which were suitable to be reintroduced into production systems.

Zero waste initiatives and circular economy dynamics help mitigating environmental concerns, by achieving innovation and ecological benefits. According to the European Parliament, slowing down the consumption of natural resources, minimizing landscape and habitat disruption, and aiding in the conservation of biodiversity are all advantages of reusing and recycling products. Moreover, adopting a circular economy approach can result in a reduction of total annual greenhouse gas emissions. According to the European Environment Agency, industrial processes and product usage contribute 9.10% to greenhouse gas emissions in the EU, while waste management accounts for 3.32% (European Parliament 2023).

In order to help mitigating and reducing paper industry contradictions with ecosystem biodiversity and maintenance of ecological integrity of natural forests, this study propose the reutilization of Invasive Alien Species *Phytolacca americana* plant waste, after cleaning initiatives, as a substitution for Eucalyptus commercial pulp in pulping and papermaking industry. The substitution of first raw materials for secondary raw matter helps mitigating not only waste production and waste burns, but reducing price volatility, availability and import dependency (European parliament 2023).

The need for creating and designing sustainable products is an increasing concern, becoming one of the most relevant issues now a days. According to the European commission Circular Economy Action Plan (2020), the anticipated global consumption of materials like biomass, fossil fuels, metals, and minerals is set to double within the next forty years. Concurrently, annual waste generation is forecasted to surge by 70% by the year 2050 (Kaza et al 2018).

For these effects, several studies have been developed to introduce *Phytolacca* fibers into laboratory paper sheets, in combination with commercial bleached pulp from Eucalyptus; physic mechanical properties has been analyzed and internal structure and chemical composition has been observed with SEM visualization. In addition, printing capacities of 100% *Phytolacca americana*, 50%-50% *Phytolacca* fiber and commercial pulp and 100% of commercial pulp has been compared, to analyze image quality obtained. Resulting materials are effectively bio composted, reducing waste production after consumption and avoiding chemical products introduction when avoiding bleaching products. The reintroduction of Invasive alien species fibers into paper production reverts not only in waste reutilization after management initiatives but wood demands from paper industry. The reduction of wood needs could be translated into the reforestation of previously monocultures areas and restoration of ecosystems, pretending biodiversity control and restructuration of affected areas.

1. Materials and methods

a. Selected species: *Phytolacca americana*

In the present study, *Phytolacca americana* (figure 1) has been selected as one of the most present species in riverbank areas and affecting considerably to riparian vegetation (Álvarez 2015). Selected area of study and recollection was the riverbank area of Umia river in Ribadumia, Galicia, Spain, which is affected with the presence of several invasive alien species due anthropogenic factors (Valero et al 2014).

Selected species *Phytolacca americana* is a perennial plant of the Polygonaceae family, native to Eastern North America, commonly known as American pokeweed, poke sallet, or inkberry. It is a large herbaceous species, reaching heights of up to 3 meters. Its stem (figure 2) is cylindrical, fleshy, and hollow inside, with purple or red spots, having a woody constitution at the base; it is easily identifiable due to its reddish/fuchsia coloration (Valero et al. 2015). The root of *Phytolacca* is thick, with multiple heads and a strong system of taproots (Balogh & Juhász 2008). The leaves are alternate, simple, lanceolate, and bi-tonal, being darker on the upper side. The maximum length of the leaves is 25 cm. Its fruit is fleshy and intensely purple, highly toxic, like all parts of the plant (Acedo & Llamas 2006).

Phytolacca americana is one of the most common herbaceous Invasive Alien Species in Mediterranean areas (Pepe et al. 2022). Nationally, it is included under Royal Decree 630/2013 in the Spanish Catalog of Invasive Alien Species; at the European level, it is regulated by Regulation (EU) 1143/2014 of the European Parliament and of the Council of 22 October 2014 on the prevention and management of the introduction and spread of Invasive Alien Species; and internationally, under the Convention on Biological Diversity (CBD) 1992, as well as the Convention on the Conservation of European Wildlife and Natural Habitats (Bern Convention, 1979) (Life, Med, WET Rivers, IAS, PHYAME/EPI/FL032).



Figure 1. (left) *Phytolacca americana* in the riverbank area of Umia river, Spain.

Figure 2. (right) Detail of stem and fruit of *Phytolacca americana*.

Currently, the distribution of this species has significantly expanded, being naturalized in various regions of the world, beyond its natural habitat in North America. From Canada to Argentina in the Americas or since the 17th century in Europe, where it began to be cultivated around the Mediterranean. References to the cultivation of this species in areas of North Africa and Southern Europe date back to the mid-17th century. Similarly, this plant has naturalized in other parts of the world, from Turkey, Asia, India, China, Taiwan, Japan, Indonesia, Australia, to New Zealand. It is one of the most widely distributed species globally and is, in numerous locations, considered an IAS (Balogh & Juhász 2008).

The plant's life cycle in the Mediterranean climate occurs between April and October, disappearing during the colder months. The plant begins to sprout as the average temperatures rise after winter, and the first plants can be found in April or May.

According to (Balogh & Juhász 2008), it can be found in uncultivated vineyards, orchards, cultivated fields, ruderal areas, or even urban areas. The fruits ripen during the warmest seasons, and it is from the last months of autumn and the first of winter that the plant disappears completely, although the root remains from one year to the next. It can be found growing annually in the same areas. In the riverbank area of Ribadumia, it can be found isolated from other individuals of its species, growing in the middle of other species such as *Tradescantia fluminensis*. But it can also be found in residual spaces between agriculture land, forming big groups of individuals, dominating the spaces.

In this study, fibers from *Phytolacca americana* were mixed with commercial bleached pulp from Eucalyptus, commonly used in paper industry. Raw matter was given to the Forestry Engineering School of University of Vigo by the ENCE Research Laboratory in Marin, Pontevedra.

b. Harvesting and delignification process

Harvesting of raw matter from *Phytolacca americana* was in June of 2022 in a specific area of 2 km in the river Umia riparian forest space, between Cabanelas and Ribadumia, Spain. Before delignification, raw matter was divided into leaves and stem, cutted in pieces of maximum 20 cm and dried in the oven at the Wood Chemistry Laboratory at the Forestry Engineering School, at a temperature of 105°C during 24h. After dehydration, stems and leaves were weighed and stored in a cooking pot of 12 L of capacity. A 20% of its weight was added in Caustic SODA (NaOH) content and distilled water was added until full covering of the material, an average quantity of 5L. As observed in previous research (Iglesias et al 2023; Kapun et al 2022) optimized times and quantities are a ratio of 5:1 liquid-to-solid mix and 90 minutes of cooking time. Temperature was selected by maintained the content at the boiling point. Internal conformation of

matter, which present strong and long fibers in the stem area, suggested the addition of 30 minutes extra for delignification process, performing a total time of 120 min at boiling temperature.

After delignification, black bleaches were filtered and stored in special containers, in order to neutralized it pH with sawdust in further occasions. As it is possible to observe in figure 3, raw delignified matter present completely different structures when referring to leaves and stem material.



Figure 3. From left to right, delignified raw matter from *Phytolacca americana* 1. Leaves, 2. Small branches and leaves mixture and 3. Stem material.

Once raw matter was cleaned with distilled water, it was dried at the oven during 24h at 105°C, for further pulping and refining processes.

Previous studies performed with *Phytolacca americana* demonstrated that more suitable results of papermaking were obtained with stem material, so in this study just stem matter was used for its blending with commercial pulp, due its similar conditions. In the publication (Iglesias et al, 2023), *Phytolacca americana* pulp material obtained a drainability degree of 63°SR and stem matter oscilated in between 21°SR and 34.5°SR in depending of refining times (60 seconds or 120 seconds). On the other hand, commercial pulp with 60 seconds of refining time obtained 27.5°SR or 43°SR for 120 seconds, with are closely to the drainability obtained with stem material. Due the finality of this study as a confirmation of the possibility of replacing commercial pulp for *Phytolacca* raw matter waste pulp, it was decided to perform tests only with stem material.

c. Pulping and refining process

Stem material from *Phytolacca americana*, once dried, was divided into small portions of 30g and rehydrated with 1L of distilled water, according to UNE-EN-ISO 5263-1:2005 indications for commercial pulp. Then, commercial pulp dried material was submitted to the same process, rehydrating it with 1L of distilled water. Refining process was performed separately.

First, unbleached matter from *Phytolacca americana* stem, was disintegrated in a Laboratory disintegration beater and obtaining a consistence of 10%, ready to be refined, following indications set up in UNE-EN ISO 4119:1996. Mixture was refined during 2 minutes in a PFI laboratory beater mill at 600 and 1200 rpm according to UNE-EN-ISO 5264-2:2011. Refined material was then weighed and distilled water was added until the obtention of 300 g of material.

Same process was performed with commercial bleached pulp from Eucalyptus. First, disintegrated and then refined for 2 minutes. Resultant pulp was measured, and matter was adjusted with distilled water until obtention of needed quantities.

In order to perform efficient laboratory sheets and physic mechanical tests, raw matter was divided in proportions of mixture of 10% - 30% - 50% - 70% -90% of raw matter from *Phytolacca* in relation with commercial pulp material. After calculation, raw matter was mixed and stored for pulping test development before laboratory sheets formation. Related information with 100% *Phytolacca americana* handsheets and

100% commercial pulp handsheet properties was used in order to have a global vision of physical characteristic evolution.

d. Physic-mechanical properties analysis of Pulp and Handsheets

After performing the refining process separately, each pulp matter was weighed to obtain the correspondent part of matter to perform each test. In order to achieve each proportion quantity for optimizing 30g of raw matter from Phytolacca and 30g of raw matter of commercial pulp following equation was calculated:

$$Q = \frac{\text{Percentage } n \times \text{raw matter mass}}{(\Sigma \text{percentage})}$$

Where:

Q is the raw matter quantity to be used.

Percentage n is the relative percentage to be calculated.

Raw matter mass is the 30g of Phytolacca or commercial pulp.

Σ percentage is the summation of all percentages to be calculated.

Result is presented as grams of mass to be used in each mixture test in order to maintain final mass constant according to norm UNE-EN-ISO 4119:1996. Proportion of distilled water added to the experiment has been adapted to maintain proportions of consistent pulp dissolution, the refining time was 120 seconds with a initial raw matter of 30 g, following pulp quantity for mixture of Phytolacca americana and Commercial pulp are presented in table 1.

Table 1. Pulp quantity to be used for mixture proportions for pulping tests.

MIXTURE PROPORTION	PULP QUANTITY FOR MIXTURE (G)	
	Phytolacca americana	Commercial pulp
10%-90%	1.2	10.8
30%-70%	3.6	8.4
50%-50%	6	6
70%-30%	8.4	3.6
90%-10%	10.8	1.2

Calculated pulp mixture was adapted to consistency needs following UNE-EN-ISO 5267-1:1999 in order to perform drainability Schopper-Riegler (°SR) test. Prepared samples of selected proportions were disintegrated and reformulated with distilled water to obtain a consistency of 0.5% to perform the test with the Schopper-Riegler apparatus.

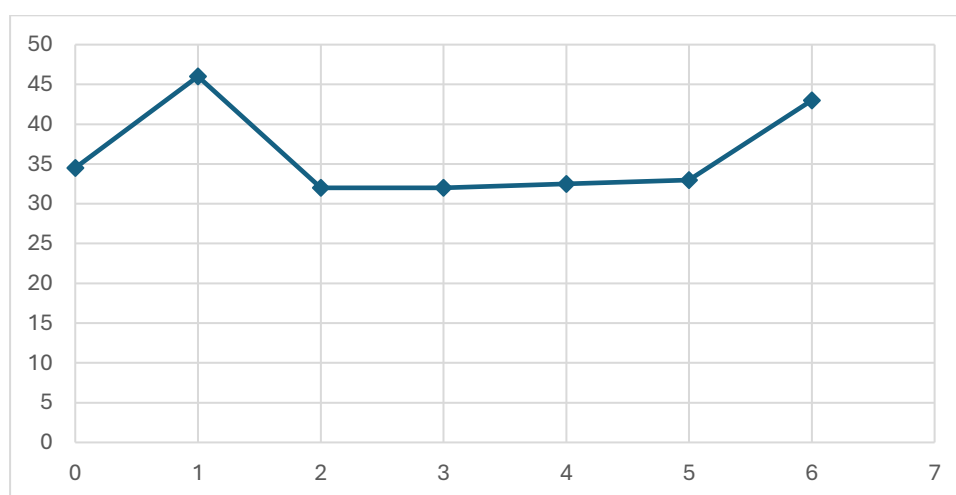


Figure 4. Drainability transformation in performed mixture test. Correspondence of Test 0=100% of Phytolacca fibers, 1= 90% of Phytolacca fibers and 10% of commercial pulp; 2= 70% - 30% ; 3= 50% - 50%; 4= 30% - 70%; 5= 10% -90% and 6= 100% of commercial pulp.

After drainability tests, laboratory sheets have been made with each pulp mixture. A conventional sheet former, ubicated at the Wood Chemistry Laboratory of the Forestry Engineering School, with a diameter of

16 cm was used with blended pulps, following UNE-EN-ISO 5269-1:2005 indications and ambient conditions were measured and set according to UNE-EN-ISO 187:1990 standards. In order to achieve an average and significant result, each test was performed in triplicates. Obtained handsheets can be observed in figure 5, where it is possible to notify color and texture variations due fiber mixture evolution.

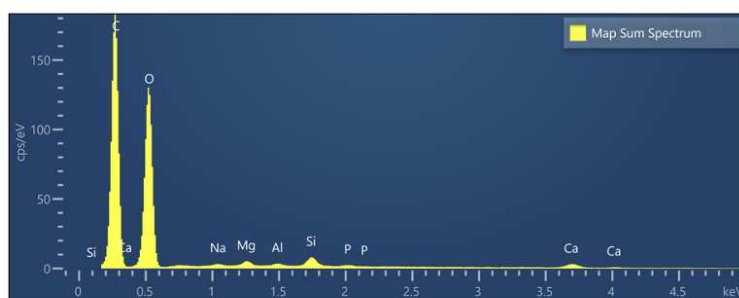


Figure 5. Laboratory sheets performed with different pulp proportions. Correspondence, from left to right: 100% of Phytolacca fibers, 90% of Phytolacca fibers and 10% of commercial pulp; 70% - 30% ; 50% - 50%; 30% - 70%; 10% -90% and 100% of commercial pulp.

With the intention to better understand paper resistance and properties, there were performed following tests to the laboratory sheets: Thickness and medium thickness (μm) and Grammage (g/m^2) according to UNE-EN ISO 536:2012; UNE-EN ISO 3039:2013; Specific volume (cm^3/g) and Specific density (g/cm^3) according to standards in UNE-EN ISO 534:2012; UNE-EN ISO 3034:2016; Air resistance was calculated with a Gurley test apparatus, expressed in second (s) and Air permeability according to Gurley ($\mu\text{m}/(\text{Pa} \cdot \text{s})$) according to standard UNE-EN ISO 5636-5:2015. All tests were performed in triplicates, following related standards.

e. Chemical composition and optical properties analysis

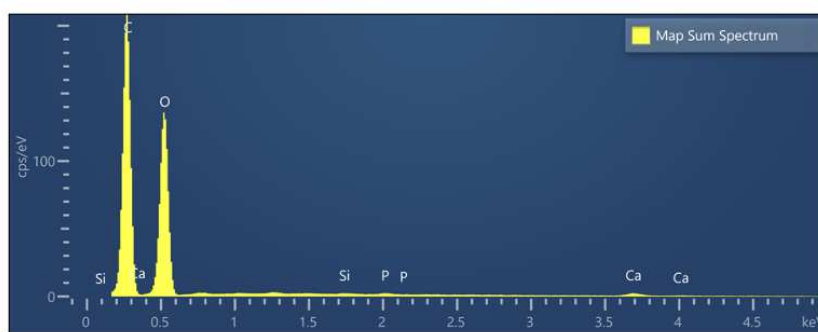
Chemical composition of performed tests was analyzed using SEM visualization at the CACTI (Help Center for Scientific and Technological Research) of the University of Vigo. Electron Microscope JEOL6100 was used in order to observe surface images of internal structure of samples and chemical composition evaluation through variations in pulp concentrations. Images of Optical microscope Nikon SZM1500 were studied to better appreciate texture and color properties of papers.



Map Sum Spectrum				
Element	Line Type	Weight %	Weight % Sigma	Atomic %
C	K series	47.93	0.16	56.15
O	K series	47.50	0.16	41.78
Mg	K series	0.57	0.03	0.33
Si	K series	1.29	0.03	0.65
Ca	K series	1.93	0.06	0.68
Na	K series	0.25	0.03	0.15
Al	K series	0.28	0.03	0.15
P	K series	0.26	0.03	0.12
Total		100.00		100.00

Figure 6. Spectrum of chemical composition of 100% commercial bleached Eucalyptus pulp, laboratory paper.

As it is possible to observe in figure 6, main composition of commercial pulp made paper is based on Carbon, Oxygen, Magnesium, Silicon, Calcium, Sodium, Alum and Phosphorus. On the contrary, 100% *Phytolacca americana* fibers made laboratory paper demonstrated a composition based on Carbon, Oxygen, Calcium, Phosphorus and Silicon (figure 7).



Map Sum Spectrum				
Element	Line Type	Weight %	Weight % Sigma	Atomic %
C	K series	49.69	0.15	57.25
O	K series	48.75	0.15	42.17
Ca	K series	1.22	0.06	0.42
P	K series	0.23	0.03	0.10
Si	K series	0.11	0.02	0.05
Total		100.00		100.00

Figure 7. Spectrum of chemical composition of 100% *Phytolacca americana* fiber's laboratory paper.

In figure 8 we observe how the internal structure of both papers present similarities, being composed by long and thin fibers that has been measured with Image J software. Fibers presented in *Phytolacca* 100% paper present an average thickness of 8.67 μm , with thinner fibers of 1.969 μm and thicker one's of 18.89 μm . Fiber length is around 200.49 μm . In comparison, fibers than form commercial pulp handsheet are an average of 10.56 μm thick, presenting more constant and stable fibers balancing between 5.099 μm and 13.99 μm . Apart from its thickness constancy, fibers are longer than 727.23 μm , being intertwined forming an homogeneous surface structure.

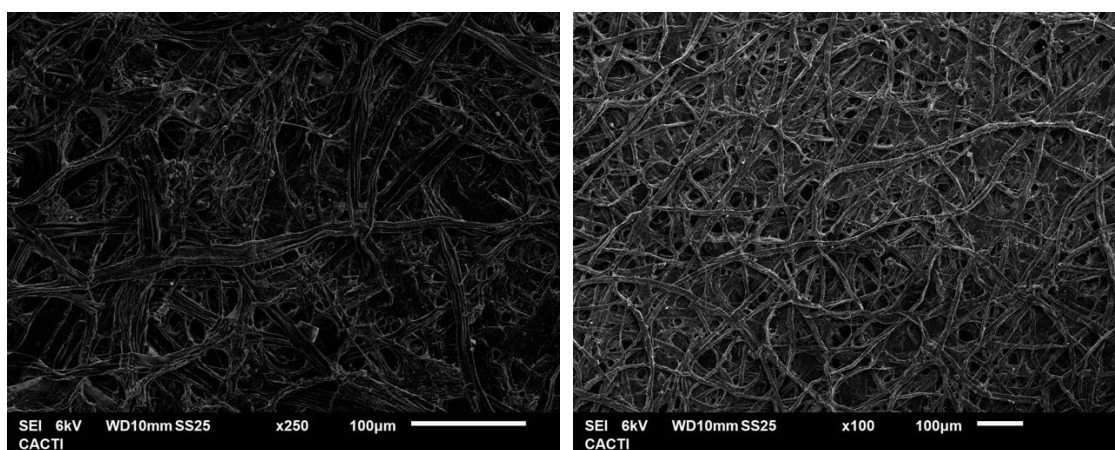


Figure 8. SEM visualization of internal structure of 100% *Phytolacca americana* laboratory paper (**right**) and SEM visualization of internal structure of 100% commercial bleached pulp handsheet. (**left**)

In figures 9, it is possible to appreciate from a lateral point of view, how papers of 100% *Phytolacca americana* (right) and 100% commercial pulp (left) are composed. As commented before, internal composition of *Phytolacca* papers are less ordinated than those made from commercial pulp.

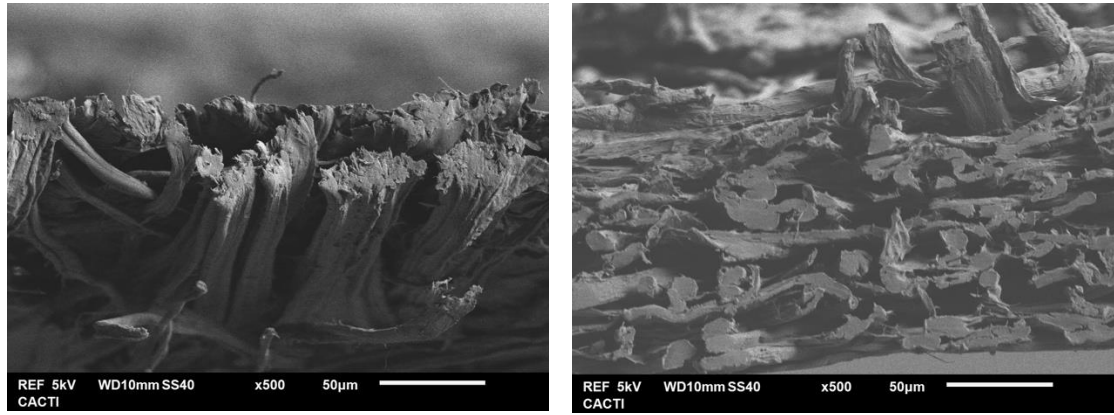


Figure 9. Transversal section of internal structure of 100% *Phytolacca americana* laboratory paper (right) and Transversal section of internal structure of 100% commercial bleached pulp handsheet (left).

f. Statistical analysis

Physic-mechanical and morphological characteristics were evaluated in accordance with mentioned ISO standards, and statistical analyses were integrated into all measurements. The properties and tests were conducted and calculated in triplicate, with the data being averaged.

Mechanical properties were tested in triplicate and carried out using PFI correspondences of 1 loop = X10 in modern equipment, were 60s was deemed equivalent to 1200 rpm (Kapun et al 2022; Iglesias et al 2023). In order to avoid misunderstandings, PFI related data is given in seconds (s) instead of rpm.

2. Results and discussion

a. Physic mechanical properties

Experiments with different proportions of pulp blend has been named as Test, corresponding Test 0: 100% *Phytolacca americana* fibers pulp; 1: 90% Phytolacca fiber 10% commercial Eucalyptus bleached pulp; 2: 70% Phytolacca fiber 30% commercial Eucalyptus bleached pulp; 3: 50% Phytolacca fiber 50% commercial Eucalyptus bleached pulp; 4: 30% Phytolacca fiber 70% commercial Eucalyptus bleached pulp; 5: 10% Phytolacca fiber 90% commercial Eucalyptus bleached pulp and 6: 100% commercial Eucalyptus bleached pulp. In following figures and tables, it will be referenced this way.

As it has been presented before, pulp drainability has been tested following Shopper-Riegler apparatus indications, obtaining an average degree of 36.14 °SR. Higher Schopper-riegler degrees are achieved in the extremes of proportions, being 100% *Phytolacca americana* and 90%-10% mixture, in addition with 100% commercial bleached pulp those with higher drainability degrees. On the contrary, even with a small descensus of drainability, it remains constant in intermediated blends, which indicates a homogeneous mixture and properties compensation of both raw materials.

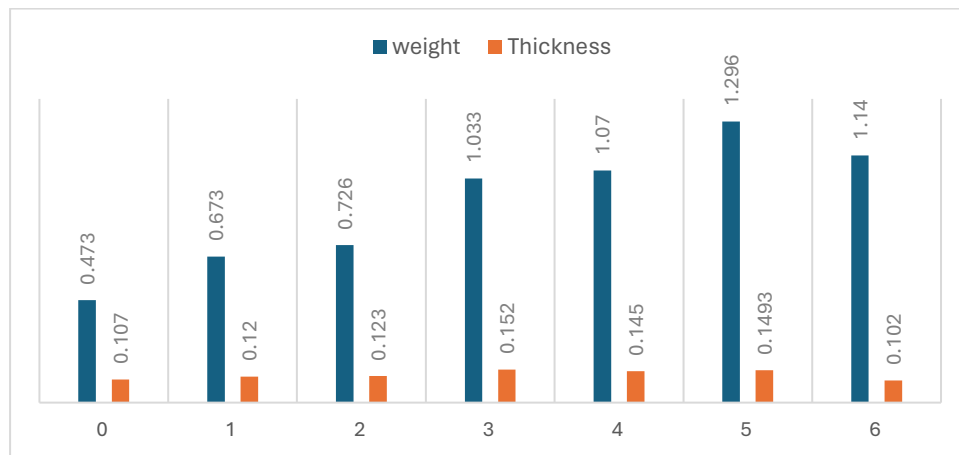


Figure 10. Graphical representation of the evolution of weight (g) and thickness (mm) in performed tests.

Physical properties relation between weight and thickness is represented in figure 10. It is possible to observe how thickness remains constant, with a variation of 0.0205 mm; but in relation with handsheet weight we can observe how commercial pulp apport heavier fibers, augmenting it as we increment commercial pulp in the mixture in decrement of *Phytolacca* fibers. Lower grammages are represented by *Phytolacca americana* 100% content and 90% with an average of 23.53 g/m² and 33.49 g/m² in front of higher grammages of tests 5 and 6 which obtain an average of 64.47 g/m² and 56.7 g/m² respectively. Medium tests with a balanced proportion of raw matters demonstrated an average of 52.39 g/m² of grammage.

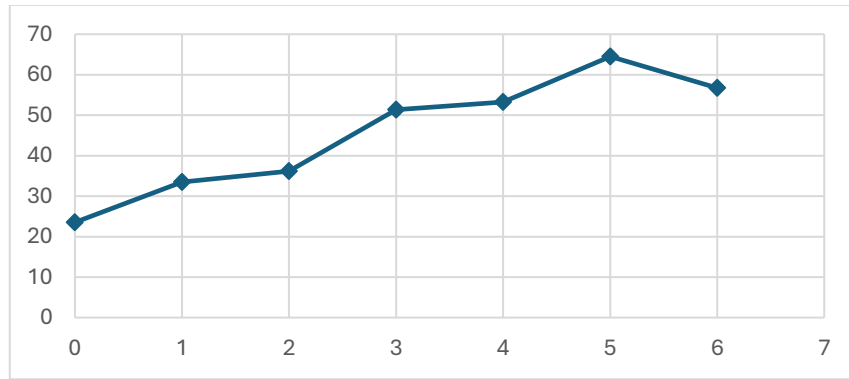


Figure 11. Evolution of grammage (g/m²) in performed tests.

In figure 11, it is possible to observe grammage evolution in performed tests, deriving to a clear augment of grammage when adding commercial pulp to the mixture. Both raw matter become integrated achieving a consistent surface. As it can be observed in table 2, in specific volume (cm³/g) and specific density (g/cm³) evolution we can appreciate same tendency, concluding higher specific density when adding commercial pulp, in decrement of specific volume (figure 12).

Table 2. Specific Volume and Specific Density tests results

TESTS	SPECIFIC VOLUME (cm ³ /g)	SPECIFIC DENSITY (G/cm ³)
0	4.56	0.22
1	3.58	0.28
2	3.41	0.29
3	2.96	0.34
4	2.73	0.37
5	2.32	0.43
6	1.79	0.56

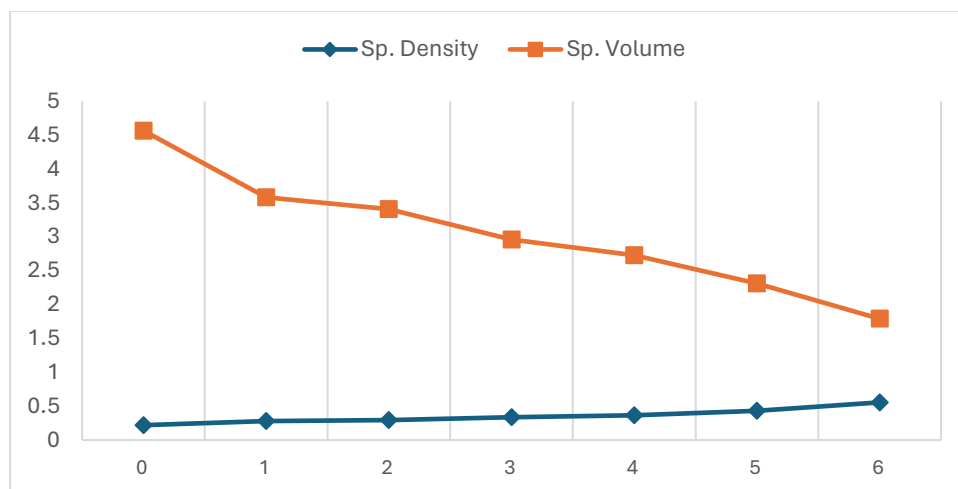


Figure 12. Inverse relation of specific density and volume along tests.

Last physic-mechanical property studied in this research was air permeability and air resistance, measured in ($\mu\text{m}/(\text{Pa} \cdot \text{s})$) and seconds (s) by using Gurley apparatus in 300 cm^3 measure. In figure 13, it is possible to observe how air permeability decrease drastically when incrementing commercial pulp to the handsheet, contrary with air resistance, which increments softly. It is possible to suppose the relation between fiber structures of different raw matter, founding a more consistent and homogeneous surface upon commercial pulp, which increments the paper capacity to resist air transmission through it. Higher air resistance times are obtained with Test 5 and 6 (9.2 s and 13.15 s) corresponding with higher contents of commercial pulp. Air permeability for these cases is the lowest obtained, being $14.7 \mu\text{m}/(\text{Pa} \cdot \text{s})$ and $10.29 \mu\text{m}/(\text{Pa} \cdot \text{s})$, respectively. In case of Tests 0 and 1 where *Phytolacca americana* concentration is higher, gurley times are reduced to 1.5 s and 1.66 s, while air permeability is $90.2 \mu\text{m}/(\text{Pa} \cdot \text{s})$ and $81.18 \mu\text{m}/(\text{Pa} \cdot \text{s})$. Medium steps, corresponding with 50% of both raw matters present an average of 2.33 seconds and $57.98 \mu\text{m}/(\text{Pa} \cdot \text{s})$.

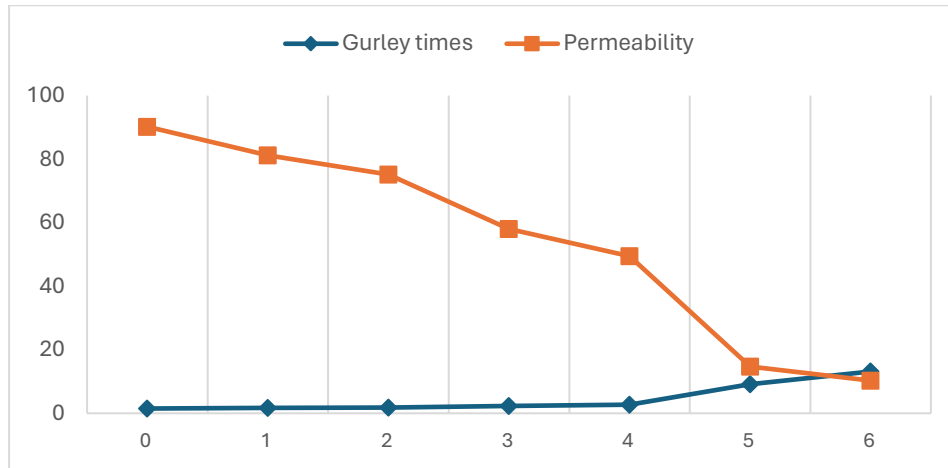


Figure 13. Air permeability ($\mu\text{m}/(\text{Pa} \cdot \text{s})$) and Air resistance (s) evolution in performed tests.

b. Internal structure integration

The internal composition of samples suffered an evolution when changing proportions of materials, but this difference is not significant when referring to surface structure due the similitudes of fibers length and thickness. In figure 14, it is possible to appreciate how main surface remains consistent, being composed by long fibers intertwined, creating a homogeneous surface in both cases. In figure 14 left, composed mainly by *Phytolacca americana* stem fibers, fibers thickness is not as consistent as in figure 14 right, but remains formatting an uniform surface.

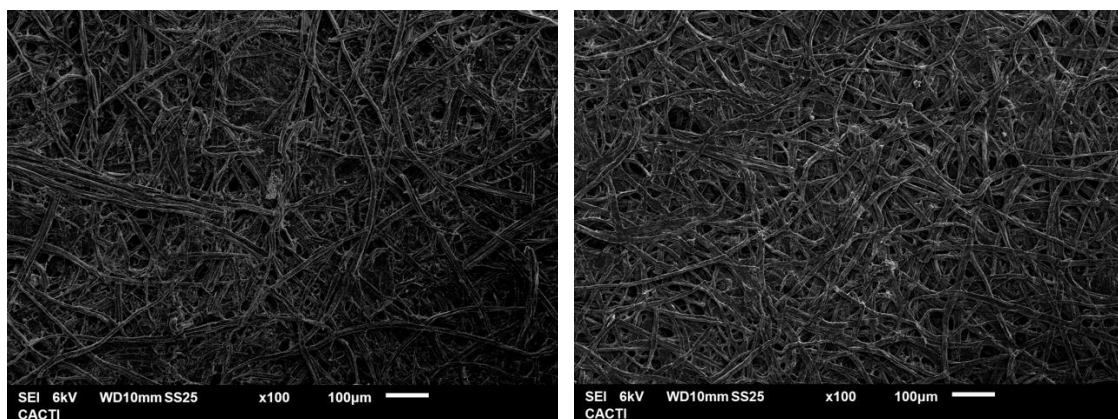


Figure 14. Electron microscope images. Internal structure of handsheet performed in test 1 (90% *Phytolacca* and 10% commercial pulp) (left) and in test 5 (10% *Phytolacca* and 90% commercial pulp) (right).

In figures 15, we observed how internal structure of proportions of test 2 and 4 remains similar while changing pulp blend. In figure 15 (left), we observe how predominant *Phytolacca*'s fibers derive in a less uniform surface, while in figure 15 (right), where there is a higher presence of commercial pulp, surface remains homogeneous. In case of intermediated compositions of test 3, 50% of both materials, it can be appreciated how the laboratory sheet remains formed by same kind of fibers (figure 16). In figure 16 right,

we observe a lateral section view of test 3 handsheet and it is possible to observe how different fibers intertwin and form a dense structure suitable to be used for any printing, writing, or cutting ends.

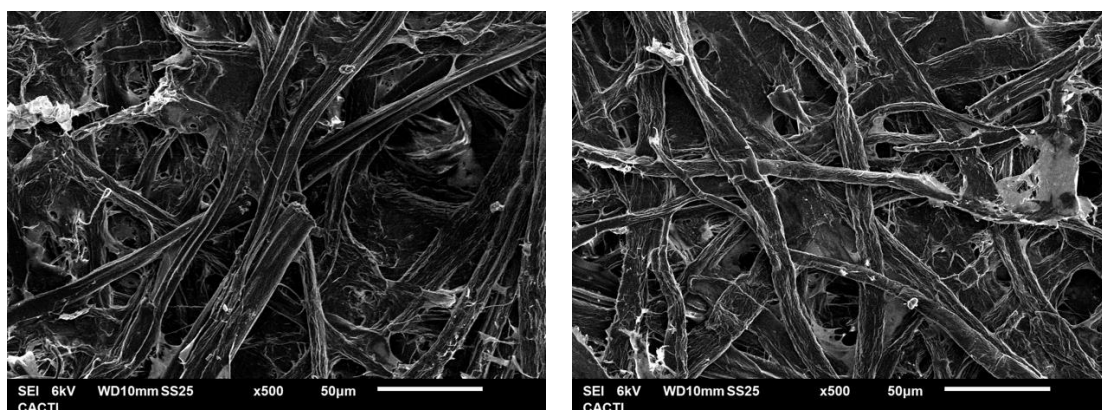


Figure 15. Electron microscope image. Internal structure of handsheet formed by 70-30% of Phytolacca and commercial pulp (left) and surface view of internal structure of 30% -70% Phytolacca material and commercial pulp (right).

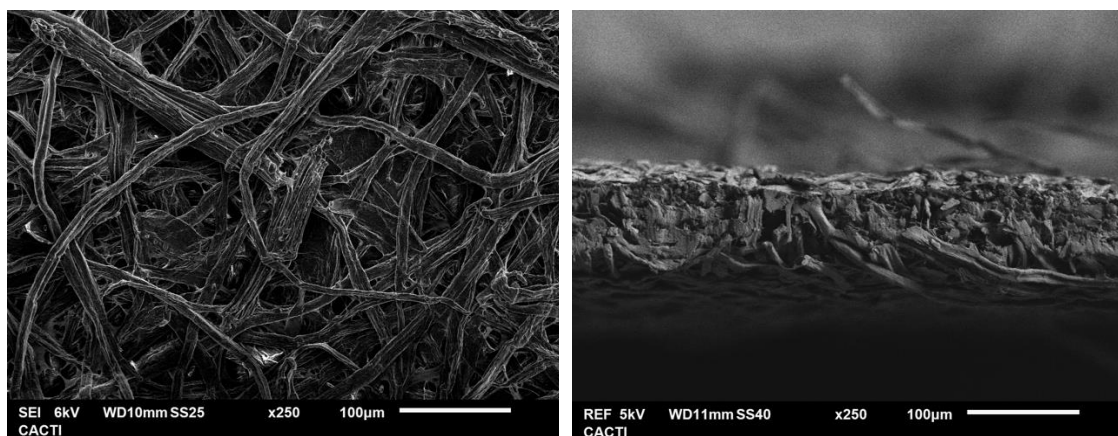


Figure 16. Electron microscope image. Internal structure of third test, formed by 50% of each material (left) and Lateral section view of internal structure of 50% of each material (right).

In figure 17, it is possible to observe how color and texture of different blends evolution when changing proportions of materials. From left to right, *Phytolacca americana* content decreases and commercial pulp increases. Yellow color present in unbleached pulp from Phytolacca gives texture and color to the laboratory sheets, adding extra value to the proposed materials.

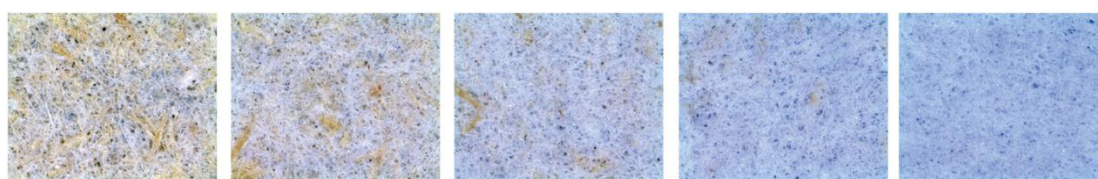
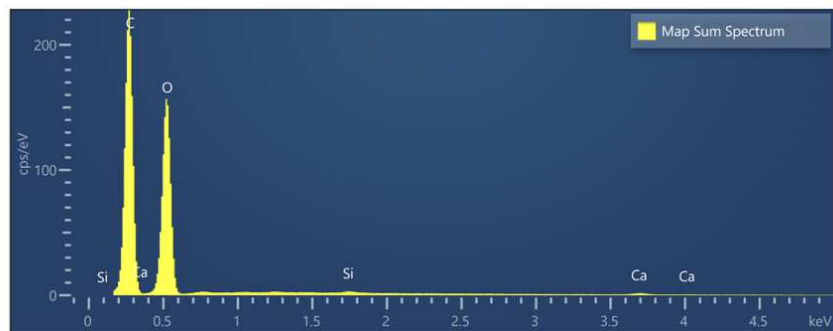


Figure 17. Color variations when changing proportions in laboratory sheets. Imagen obtained from optical microscope Nikon SZM1500.

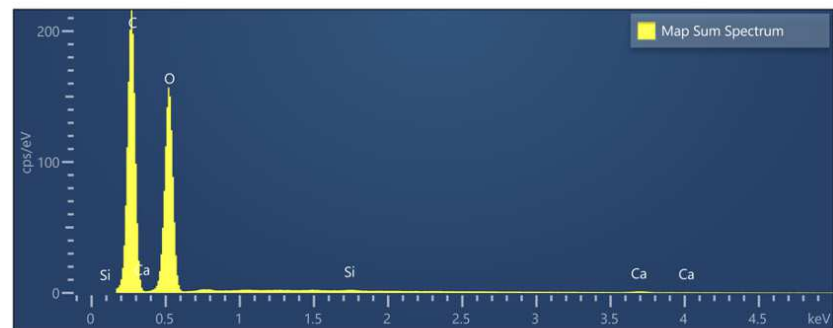
c. Chemical composition evolution

Internal composition of samples has been studied with SEM visualization. As it has been observed, main composition of both materials demonstrated Carbon and Oxygen as principal components. Differing from commercial pulp paper, *Phytolacca americana* composition present less chemical compounds as it is not bleached, and this variation can be observed in figures 18 and 19, representing test 2 and 4 (70% Phytolacca and 30% commercial pulp - 30% Phytolacca and 70% commercial pulp).



Map Sum Spectrum				
Element	Line Type	Weight %	Weight % Sigma	Atomic %
C	K series	49.16	0.15	56.53
O	K series	49.98	0.15	43.15
Ca	K series	0.68	0.05	0.23
Si	K series	0.18	0.02	0.09
Total		100.00		100.00

Figure 18. Spectrum of composition in test 2.



Map Sum Spectrum				
Element	Line Type	Weight %	Weight % Sigma	Atomic %
C	K series	48.57	0.12	55.86
O	K series	50.88	0.12	43.93
Ca	K series	0.44	0.04	0.15
Si	K series	0.11	0.02	0.06
Total		100.00		100.00

Figure 19. Spectrum of composition in test 4.

As we observe in figures 18 and 19, composition remains almost constant in both tests, and in figure 20 and 21 we can appreciate the slight evolution when mixing both materials along different tests. Main components remain Carbon and Oxygen, which exchange quantities after test 2, when major content of Oxygen is presence in decrement of Carbon. Higher proportion of commercial pulp derives to higher content of Oxygen. Contrary, higher content of Phytolacca material leads to higher Carbon proportion in mixture. In figure 19 we can appreciate how Calcium and Silicon, both secondary most present components evolution when changing proportions of Phytolacca and commercial pulp. Both components percentage tends to 0 when augmenting commercial pulp material. Silicon disappears in test 5 when Phytolacca mater is just 10% of mixture.

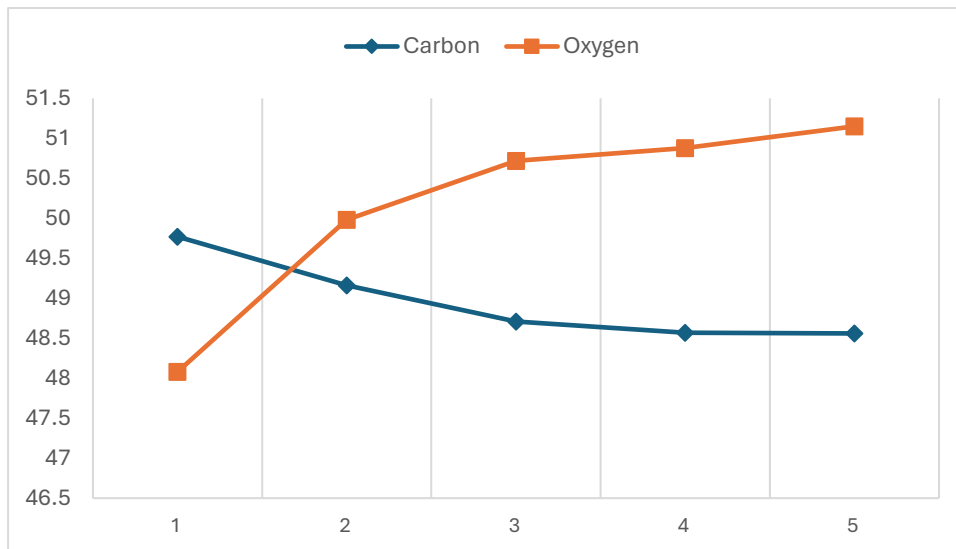


Figure 20. Evolution of percentage of Carbon and Oxygen content when changing proportions of raw matter.

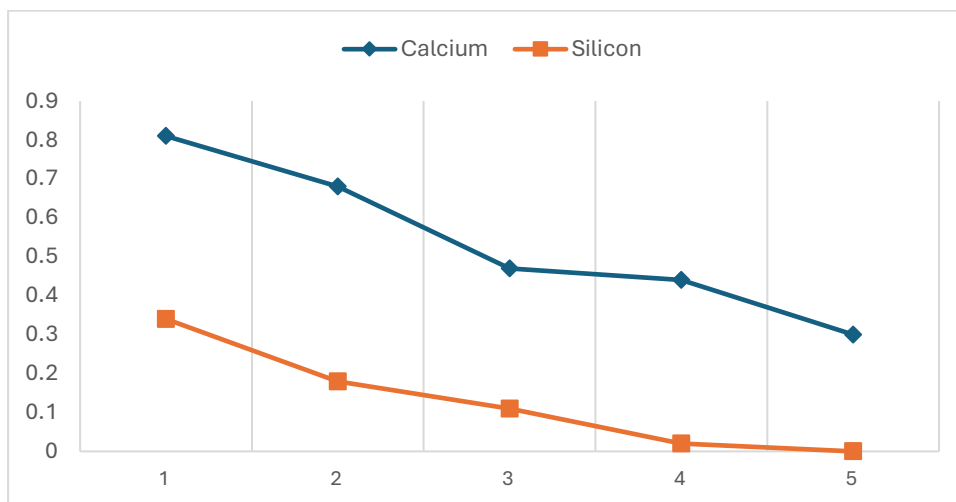


Figure 21. Evolution of percentage of Calcium and Silicon content when changing proportions of raw matter.

As it has been demonstrated, physic mechanical, chemical and optical properties of developed handsheets suffered evolutions when changing materials proportions. But it is relevant to mention that difference doesn't reflect on quality or resistance of material, concluding that its blend is suitable to be applied in industry.

When referring to physical properties, higher grammages are obtained with higher commercial pulp content in relation with *Phytolacca americana* material but it remains higher with mixture of 10%-90% of Phytolacca and commercial pulp, than with 100% of commercial pulp. Furthermore, paper specific volume has been incremented when adding higher quantities of Phytolacca, and air resistance keeps constant when mixing material in same proportions. Regarding chemical composition, there are no significant changes that reflects con bio composability of the material; and in relation with optical properties, adding *Phytolacca americana* to commercial bleached eucalyptus pulp proposed value added results, obtaining textures and colors suitable to be of interest in creative and packaging industries.

3. Conclusion

Conducted research on the utilization of *Phytolacca americana* fibers in combination with commercial pulp material has demonstrated significant potential for paper industry. Value added products demonstrates great possibilities for artistic applications and environmental communication purposes as it offers a variety of color and texturized papers along proportions variations. The experiments undertaken have successfully optimized blending ratios, showcasing the feasibility of incorporating waste biomass from invasive alien species *Phytolacca americana* into paper production. This approach not only promotes circular economy

principles but also contributes to waste reduction initiatives, thereby mitigating the need for organic waste incineration.

The developed paper exhibits promising optical and structural properties, with varying proportions yielding diverse physical and mechanical characteristics suitable for a range of applications. Intermediated proportions of 50% of both materials demonstrating maintaining initial properties of commercial pulp paper with added values of color, texture, and fiber reutilization. Presented results confirm the feasible possibility of replacing commercial bleached pulp for proposed vegetal waste material without variation of properties, neither chemical, physic-mechanical nor optical. By harnessing waste biomass, particularly from invasive alien plant species, sustainable papermaking practices are promoted, aligning with biodiversity conservation goals and environmental stewardship principles. This innovative approach offers an eco-friendly alternative, addressing pressing environmental challenges within the papermaking sector, contributing to the possibility of reduction of wood demand for paper industry, which will relay into monocultures reduction and local biodiversity maintenance.

As the industry faces increasing environmental pressures, embracing innovative and sustainable methodologies is imperative for reducing ecological footprints. By promoting the adoption of such approaches, the papermaking sector can transition towards a more environmentally responsible and resource-efficient future, contributing to global efforts to combat climate change and preserve biodiversity. Simultaneously, a new paper surface with great printing and decorative ends is proposed for the creative industry, opening a huge range of possibilities in order to embrace environmental awareness and care.

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