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Research Article

Development and Characterization of a Biodegradable pH Indicator Film Using Plant-Based Pigments

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Abstract

Natural pigments derived from plants present a promising alternative to synthetic dyes for use as pH indicators, owing to their sustainable nature and minimal environmental impact. This study investigates the extraction and characterization of plant-based pigments to create pH-sensitive paper materials. Unlike synthetic pH indicators, which contribute to chemical waste and environmental pollution, these biodegradable alternatives offer an eco-friendly solution for pH testing. The work outlines a systematic approach to evaluate the color changes of plant pigments when exposed to various pH levels while simultaneously developing biological pH-dependent papers and assessing their potential applications as chemical sensing devices. Pigments were extracted from the flowers of *Cosmos bipinnatus*, *Impatiens balsamina*, *Tabernaemontana divaricata*, and *Tagetes erecta*. The paper substrates were composed of fibers from Panicum virgatum (switchgrass). Color transitions were analyzed using pH meter calibration and UV-visible spectroscopic techniques. Results indicated that the floral pigments in *Impatiens balsamina* exhibited significant color variation with pH due to anthocyanin content, while the pigments from *Tagetes erecta* demonstrated stability attributed to carotenoids. The resulting paper displayed strong pH sensitivity, confirming its viability as a pH detection tool. The findings support the potential of plant-based pigments for developing responsive pH paper. Future work should focus on enhancing pigment longevity, scaling production, and exploring practical applications in environmental monitoring, food preservation, and medical diagnostics.

Keywords: Anthocyanins; pH-Sensitive Paper; Synthetic Dyes; Switch Grass (*Panicum Virgatum*); Spectrometer.

1. INTRODUCTION

In addition to their health benefits, natural pigments contribute to sustainable practices by providing eco-friendly alternatives to synthetic dyes, thereby reducing chemical waste and promoting biodiversity through conservation efforts [12], [13]. Carotenoids, prominent in many vegetables and fruits, play a vital role in human health by supporting vision and immune function and have potential applications in food preservation due to their natural antioxidant properties [14]–[16]. These natural pigments' multifaceted applications and advantages underscore the importance of exploring and preserving the rich plant diversity in regions like Sri Lanka. Moreover, as the demand for sustainable and health-conscious products increases, the continued research and development of natural pigments will play a crucial role in various industries, including food, cosmetics, and pharmaceuticals [11], [17].

Plant pigments, such as chlorophyll, are essential for processes like photosynthesis, converting sunlight into energy, while carotenoids (e.g., lutein in *Tagetes erecta*) enhance light absorption and protect against photo-oxidative damage [18], [19]. Brightly colored flowers, including *Clitoria ternatea* and *Cosmos bipinnatus*, attract pollinators through anthocyanins and carotenoids, ensuring reproductive success [20]. Moreover, flavonoids and anthocyanins absorb harmful ultraviolet radiation, providing crucial DNA protection; for instance, *Tabernaemontana divaricata* absorbs indole alkaloids and flavonoids to enhance UV resistance [21].

Modern industries harness these pigments for innovative uses, blending traditional practices with contemporary applications. In pharmaceuticals, anthocyanins from *Clitoria ternatea* offer antioxidant properties for cognitive health [22], [23], while *Tagetes erecta* provides lutein, which is beneficial for eye health and as a natural food colorant [24], [25]. In food, butterfly pea flower extract is used for its pH-sensitive color-changing properties [26]–[28]. The cosmetics industry utilizes natural Curcuma Longa and Bixa Orellana dyes, replacing harmful synthetic dyes [29], [30]. Additionally, Sri Lanka's batik and handloom industries utilize plant-derived pigments, reducing reliance on toxic synthetic dyes [31].

Despite these advantages, natural pigments face stability challenges under heat, light, and pH changes [32], [33]. The research addresses these issues through encapsulation techniques to prolong shelf life and employs nanotechnology to stabilize carotenoids in cosmetics. Promoting sustainable agroforestry practices enables the cultivation of pigment-rich plants, such as Clitoria ternatea, while preserving biodiversity. Through these efforts, the potential of Sri Lanka's natural pigments is being expanded, supporting both ecological sustainability and industrial innovation.

The study of natural pH indicators has played a pivotal role in the development of chemistry, tracing its origins to the 17th century. Robert Boyle, often regarded as the father of modern chemistry, pioneered this field by documenting the color-changing properties of plant extracts in response to acidic or alkaline environments. In his seminal work, The Experimental History of Colors (1664), Boyle described how violet syrup derived from plants like violets and cornflowers shifted hues when exposed to acids (e.g., vinegar) or bases (e.g., limewater). This discovery laid the groundwork for understanding the relationship between chemical reactivity and visual indicators [34].

In the 20th century, advancements in organic chemistry deepened our understanding of natural dyes. Nobel laureate Richard Martin Willstätter's research on plant pigments, particularly chlorophyll and anthocyanins, revealed the structural basis for their pH sensitivity. Willstätter demonstrated that anthocyanins—flavonoid compounds responsible for red, blue, and purple hues in flowers and fruits—undergo structural rearrangements in varying pH conditions, altering their light absorption spectra and thus their visible color [35], [36]. Later, researchers such as Helmut Schweppe and Franco Brunello expanded on these findings, cataloging plant sources like litmus (from lichens) and curcumin (from turmeric) and their applications in analytical chemistry. Handbook of Natural Dyes [37] became a key reference for identifying pH-sensitive pigments in flora. At the same time, Brunello emphasized the plant-based dyes' cultural and industrial significance in The Art of Dyeing in the History of Mankind [38].

Modern studies highlight anthocyanins as exceptionally versatile natural pH indicators. These water-soluble pigments, abundant in sources like red cabbage (*Brassica oleracea*), blueberries (*Vaccinium corymbosum*), and hibiscus flowers (*Hibiscus sabdariffa*), exhibit vivid color transitions across a broad pH range. For instance, red cabbage anthocyanins shift from red (pH $\leq$ 3) to purple (pH 7–8) and greenish-yellow (pH $\geq$ 11) due to protonation/deprotonation of hydroxyl groups on their flavylium cation structure [39]–[41]. Recent research has optimized their extraction and stabilization, addressing challenges like thermal degradation and light sensitivity. Techniques such as encapsulation with biopolymers (e.g., chitosan) or nanocomposites have enhanced their durability for commercial use [42], [43].

This study has broad significance in several aspects. From an environmental perspective, replacing synthetic pH indicators with natural pigments could reduce the impact of chemical waste on pollution. Industrially, the findings of this research could be applied in pharmaceuticals, food production, and textile manufacturing, which are increasingly shifting towards natural materials. Scientifically, this study contributes to a deeper understanding of the stability of natural pigments and their potential in analytical applications. Economically, utilizing local plant resources could promote a more sustainable and economically viable approach to biodiversity utilization.

Several research gaps have been identified in this field. One of the primary challenges is enhancing the stability of natural pigments against environmental factors, including light, temperature, and extreme pH conditions. Another significant gap involves identifying the most efficient extraction methods for high-purity and efficient pigments. Additionally, integrating natural pigments into a pH-responsive paper system that can be widely applied for pH testing remains an open challenge.

The primary objective of this research is to develop a natural pigment-based pH indicator system derived from flowers, fruits, and tubers that can exhibit color changes in response to pH variations. Specifically, this study aims to identify plant sources with highly pH-sensitive pigments, evaluate extraction and stabilization methods for pH indicator applications, develop a prototype of a pH-responsive paper based on natural pigments that are effective and environmentally friendly, and analyze the stability of natural pigments under various environmental conditions to ensure long-term applicability.

2. MATERIALS & METHODS

2.1 Preparation of Extract

The materials used for the preparation of extracts include various flowers from green plants: Tugger flower (*Tabernaemontana divaricata*), Yellow Cosmos flower (*Cosmos bipinnatus*), Marigold flower (*Tagetes erecta*), Orange Rubiaceous flower (*Ixora coccinea*), Red Kudalu and Pink Kudalu flowers (*Impatiens balsamina*), Periwinkle flower (*Catharanthus roseus*), Ground Orchid flower (*Spathoglottis plicata*), Butterfly pea flower (*Clitoria ternatea*), Anthurium Green (*Anthurium andraeanum*), as well as vegetables and fruits such as Purple potato (*Dioscorea alata*), Carrot (*Daucus carota*), Beet (*Beta vulgaris*), Turmeric (*Curcuma longa*), and

Bael (*Aegle marmelos*). These materials were sourced from gardens in Maharagama and Kandy, Sri Lanka.

To prepare the flower extracts, the petals of each flower were collected and rinsed thoroughly with distilled water. They were then pressed between pads of tissue paper to remove excess surface water. Fresh petals (5g) of each flower were placed in a pestle and chopped with a mortar. Distilled water (10 mL) was added to the pestle, and the mixture was thoroughly blended. The resulting extracts were filtered using filter paper and transferred into a 50 ml beaker with a pipette. An additional 20 ml of distilled water was added to each beaker to dilute the flower extracts.

Similarly, the potato and fruit extracts were rinsed with distilled water, and excess water was removed using tissue paper. The potatoes and fruits were cut into small pieces and chopped to facilitate extraction. Distilled water (10 mL) was then added to the pestle, and the mixture was filtered using filter paper before being transferred to a 50 mL beaker using a pipette. Again, 20 ml of distilled water was added to the beakers to dilute the extracts from the potatoes and fruits.

Figure 1. Materials to Prepare Flower Extract

Paper chromatography was used to identify plant pigments in the various flowers. Paper Chromatography is a chemical purification method that separates colored substances. In paper chromatography, pigments can be separated based on the differences in the sizes of their molecules. Leaves contain chlorophyll, which is green, but plants contain a wide range of other pigment molecules. Plant cells are broken open for paper chromatography to release their pigment molecules. A solution of plant matter and alcohol is placed at the bottom of a paper. Alcohol moves up the paper, taking pigment molecules with it. It is easier for smaller molecules to move through the fibers in paper, so they travel fastest and move the furthest up the paper. Larger molecules are slower and do not travel as far up the paper.

2.2 Determination of Color Change in Different pH

This study investigates the color changes in various natural extracts in response to different pH levels. The materials utilized included acidic solutions, such as battery water containing Sulphury acid (H_2SO_4) and vinegar (acetic acid, CH_3COOH), along with essential solutions like caustic soda (sodium hydroxide, NaOH) and baking soda (sodium bicarbonate, $NaHCO_3$). Flower, potato, and fruit extracts were selected for their potential as natural pH indicators.

The experiment began with labeling 50 mL volumetric flasks and adding 30 mL of each extract. Subsequently, titration was performed using a burette to introduce either Sulphuric acid or Acetic acid, followed by Sodium bicarbonate, until neutralization was achieved. The pH values were recorded using a calibrated pH meter, providing insight into the acidity or basicity of the solutions.

UV-visible spectroscopy was employed to analyze color changes quantitatively. The experiment was repeated for all extracts to ensure reliability and consistency in the results. This systematic approach demonstrated the capability of natural extracts to act as effective pH indicators, contributing to a deeper understanding of the relationship between pH levels and colorimetric changes.

2.2.1. pH Meter Method

The experiment commenced with the calibration of the pH meter. The protective cap was removed from the electrode and rinsed with distilled water. Calibration began with a buffer solution of pH 7; the 'CAL' button was pressed before submerging the electrode. The pH meter was calibrated when the readings matched the standardized value of the buffer. After this, the electrode was rinsed again with distilled water. The process was repeated using buffer solutions of pH four and 10, ensuring the electrode was rinsed after each use and the protective cap was reattached when the calibration process was paused.

For the measurement of neutralized solutions, prepared from the titration of acids and bases into flower extracts, the solutions were placed under the electrode of the stationary pH meter. The electrode was carefully submerged in the neutralized solution and readings were recorded. After measurement, the electrode was rinsed with distilled water, and the protective cap was reattached.

2.2.2. UV-Visible Spectroscopy Method

The UV-visible spectrophotometer was turned on, and a warm-up period of at least 20 minutes was allowed. The absorption spectrum was determined using a standard acetate buffer as the blank solution. Care was taken to select cuvettes specifically for this purpose, ensuring that the lower portion, which allows light to pass through, was not handled. The cuvettes were rinsed multiple times with the acetate buffer solution before inserting the blank cuvette into the cell holder, with the index line facing forward to avoid scratches. The wavelength control knob was set to 265 nm for calibration, allowing observation of pigment variations at different pH levels. The outer surfaces of the cuvettes were wiped with tissue paper to ensure accuracy in measurements.

2.3 Process of Paper Making

The initial step involved cutting the harvested switch grass into small pieces, approximately 2-3 cm long, which were soaked in distilled water overnight. This soaking period allowed the fibers to soften, preparing them for further processing. The next day, the soaked grass was transferred to a large stainless steel pot and cooked for approximately three hours. This cooking process effectively broke down the plant material into pulp, which was then strained to separate the liquid from the softened fibers.

Figure 2. Process of Creating Paper from Switch Grass (*Panicum virgatum*)

Once the fibers were prepared, they were blended into a smooth pulp. Isopropyl alcohol was added to the mixture to enhance the paper's quality and remove any residual chlorophyll. The resulting pulp was poured into a large plastic box and shaped using a mound with the screen side facing up, topped with a deckle to define the edges. The pulp was pressed onto the mound, allowing excess moisture to drain, and absorbent cloths were used to couch the paper. Finally, the newly formed sheets were placed on boards to dry flat, transforming switch grass into functional paper. This method exemplifies a sustainable approach to paper production, effectively utilizing natural materials.

2.3.1. Chlorophyll Removal Method

This study employs an effective method for chlorophyll removal, utilizing alcohol as the solvent. The necessary materials include leaf pulp, isopropyl alcohol, ethyl alcohol (EtOH), or rubbing alcohol, as well as a medium-sized pot and a sieve.

The procedure begins by pouring isopropyl alcohol, ethyl alcohol, or rubbing alcohol into a medium-sized pot. Subsequently, the leaf pulp is added to the pot, ensuring that it is fully submerged in the alcohol. This mixture is allowed to sit for 24 hours to facilitate optimal chlorophyll extraction. Following this period, the leaf pulp is filtered using a sieve, separating the liquid containing the extracted chlorophyll. This filtration process is repeated twice, using fresh alcohol each time to ensure thorough and effective chlorophyll removal.

2.4 Dyeing of Paper from Plant Pigments to Produce Final Product as pH Indicator Film

The process of dyeing paper using plant pigments is explored to create a final product that serves as a pH indicator film. Several methods are employed in this endeavor, including Solution Dyeing, Coloration, and Absorption.

2.4.1. Solution Dyeing

The first method utilized is known as solution dyeing, also called dope dyeing or spun dyeing. In this process, the pigment color is bonded within a solution and is absorbed by the fibers as they are formed in the dye liquor. This approach is practical for cellulosic and non-cellulosic fibers, producing vibrant, clear, and long-lasting colors. The materials needed for this method include biological pigments or dyes, prepared paper, water, and a medium-sized pot.

The methodology begins with dyeing the fibers in an aqueous solution called the dye liquor or bath. For dyeing to be successful, the fibers must undergo a thorough coloration process and effective dye absorption.

2.4.2. Coloration

Coloration plays a crucial role in the dyeing process, as it determines both the aesthetic appeal and durability of the dyed material [44], [45]. The primary objective is to achieve a stable and long-lasting coloration that remains intact even after repeated exposure to water or regular washing [46]. Moreover, the dye must exhibit strong resistance to fading when exposed to light, ensuring that the vibrancy and intensity of the colors are preserved over time. An ideal dye should also demonstrate high affinity to the substrate, allowing for uniform absorption and optimal fixation, which contribute to enhanced color fastness and overall durability.

2.4.3. Absorption

The absorption of the dye molecules into the fiber is the final component in this process. As the dye molecules concentrate on the fiber surface, they become securely attached through various forces. These include ionic forces, hydrogen bonding, Van der Waals, and covalent chemical linkages. The interplay of these interactions is crucial for enabling the dye to adhere effectively to the paper, resulting in a stable and functional pH indicator film. Through the careful application of these dyeing methods, this study successfully demonstrates the creation of paper dyed with plant pigments, resulting in a reliable product for use as a pH indicator film.

3. RESULTS

4.1. Paper Making

This study demonstrates the process of making paper from switchgrass (Panicum virgatum) plant fibers. Converting plant fibers into paper involves five consecutive stages: material collection, preparation, pulping, molding, and drying. Each stage contributes to the final product's quality characteristics, texture, and effectiveness. Researchers selected switchgrass as the raw material due to its high cellulose fiber content, which allows for the production of durable paper.

During this process, organisms containing chlorophyll exhibit a green color, necessitating additional steps to remove chlorophyll to prevent decomposition and color imbalance in the paper. The selection of plant materials is crucial in determining the resulting paper's strength, flexibility, and porosity. Switchgrass is suitable because of its strong fibers and highly renewable properties.

Researchers precisely cut switchgrass leaves using small-scale cutting methods. Reducing plant material size increases the surface area exposed to processing, accelerating fiber breakdown. Subsequent steps benefit from improved water and alcohol penetration, making fiber extraction easier. Cutting the material to the appropriate size enhances lignin removal, expediting the pulping process. Soaking the plant fibers initiates lignin degradation, facilitating better fiber separation. This process also promotes the initial release of pigment, contributing to improved color uniformity in the paper. Pre-treating the plant fibers enhances the efficiency of subsequent processing, particularly during blending.

During pulp production, adjustments in blending or thickening are necessary if the pulp becomes too watery or lumpy. The texture, thickness, and absorption capacity of the final paper depends entirely on the fiber standards. The molding process shapes the paper sheet as it drains excess water. Even the distribution of pulp ensures consistent thickness and a stable structural framework in the finished paper. The molding stage determines the paper's surface characteristics, ultimately influencing its usability as either a writing material or a pH indicator.

The study results indicate that switchgrass is suitable for plant-based paper production. The chlorophyll content in plant fibers requires alcohol soaking to prevent degradation, which can impact the paper's color. Proper fiber blending and soaking sessions produce a more evenly distributed pulp. However, excessive processing weakens the fibers, while inadequate processing results in uneven paper formation. Evenly formed and uniformly dried paper sheets achieve optimal strength and flexibility, with their natural fiber arrangement defining absorbency, strength, and usability properties. Applying plant-based colorants to the paper enables its transformation into pH indicator films, which are helpful for chemical testing.

Table 2. Color Change with Different pH Levels

4.2. Universal pH Indicator

A universal indicator is a combination of pH indicator solutions that can be used to determine the pH of a solution over a wide range of values. There are numerous formulas for universal indicators. Standard mixture components include Thymol blue, methyl red, bromothymol blue, and phenolphthalein. The color change is used to identify pH values. The following are the most common universal indicator colors:

Table 1. Indicator Color and pH Values

Indicator Colors	pH Values
Red	$0 \geq pH \geq 3$
Yellow	$3 \geq pH \geq 6$
Green	$pH = 7$
Blue	$8 \geq pH \geq 11$
Purple	$11 \geq pH \geq 14$

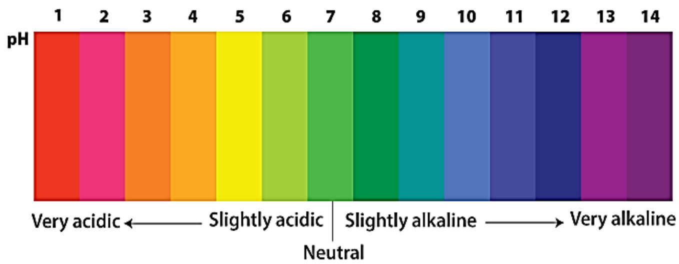

Figure 3. Color Variation in Different pH Scales and Indicators

pH indicators are vital tools in chemistry for estimating acidity or alkalinity in various solutions, with their primary role being to signal the completion of acid-base reactions during titrations. While pH meters provide precise measurements, indicators are helpful for quick assessments in diverse applications such as measuring farm soil pH for optimal plant growth, checking the acidity of shampoos for hair suitability, assessing the pH of fruit juices for flavor and preservation, and monitoring the health of bodies of water to maintain ecological balance.

Name of the Flower	Previous Color	Color Change After Adding		Descriptions
		Base	Acid	
Tugger Flower (*Tabernaemontana Divaricate*)	White	Light Yellow	Not Changed	
Yellow Cosmos Flower - *Cosmos Bipinnatus*	Light Yellow	Light Yellow	Light Yellow	
Marigold Flower (*Tagetes Erecta*)	Orange	Yellow	Not Changed	
Orange Rubiaceous Flower (*Ixora coccinea*)	Light Orange/ Creamish Pink	Not Changed	Not Changed	
Red Kudalu Flowers (*Impatiens balsamina*)	Red	Brick Red	Light Red	
Pink Kudalu Flowers (*Impatiens balsamina*)	Pink	Yellow color	Light Purple	
Periwinkle Flower (*Catharanthus Roseus*)	Purple	Green	Pink	
Ground Orchid Flower (*Spathoglottis plicata*)	Light Purple	light green	Not changed	
Butterfly Pea Flower (*Clitoria Ternatea*)	Blue	Bluish-Green	Dark purple	
Purple potato (*Dioscorea Alata*)	Purple	Deep Blue	Deep Pink	
Carrot (*Daucus Carota*)	Orange	Yellowish	Not Change	

Name of the Flower	Previous Color	Color Change After Adding		Descriptions
		Base	Acid	
Beet (*Beta Vulgaris*)	Dark Purple	Dark Purple	Light Purple	
Turmeric (*Curcuma Longa)*	Yellow	Light Red	Not Changed	
Bael (Beli) Fruit *(Aegle Marmelos)*	Light Yellow	Not Changed	Not Changed	

Anthocyanin pigments in various flowers, such as purple potatoes (*Dioscorea alata*) and periwinkle flowers (*Catharanthus roseus*), exhibit color variations in response to changes in environmental pH. These pigments, which contribute to blue, purple, and red hues in plants, appear red or purple under acidic conditions but shift to blue or green in alkaline environments. The butterfly pea flower (*Clitoria ternatea*) exemplifies this transformation, as exposure to alkaline solutions alters its color from blue to a deep purple. This phenomenon highlights how anthocyanins alter their light absorption properties in response to pH fluctuations.

Under acidic conditions, the yellow Cosmos flower (*Cosmos bipinnatus*) undergoes a color change to brick-red, suggesting a chemical transformation of its yellow flavonoid pigments. Conversely, when exposed to a basic solution, the flower exhibits a lighter yellow hue, indicating possible pigment neutralization or structural degradation. Similarly, the pink Kudalu flower (*Impatiens balsamina*) transitions from pink to yellow in acidic environments and shifts to purple under alkaline conditions. These color transformations occur due to the protonation or deprotonation of anthocyanins in response to varying pH levels.

In contrast, the pigments in the Tugger flower (*Tabernaemontana d*ivaricata) and the orange Rubiaceous flower (*Ixora coccinea*) remain unaffected by acid or base solutions, indicating their pH stability. This resistance to pH changes suggests that their pigmentation is primarily derived from carotenoids, which are known for their chemical stability in the face of pH variations. Similarly, the marigold flower (*Tagetes erecta*) retains its color under acidic conditions but turns yellow in basic environments. The yellow coloration in marigolds originates from carotenoids, which may undergo structural modifications under alkaline conditions.

The betacyanin pigments in Beta vulgaris remain stable in acidic solutions but exhibit a lighter purple shade when exposed to alkaline environments. This response suggests that betacyanins demonstrate moderate sensitivity to basic conditions while maintaining stability in acidic media. Meanwhile, the curcuminoid compounds in turmeric (*Curcuma longa*) remain stable across different pH levels; however, they turn a light red under acidic conditions. This color shift likely results from protonation reactions that alter the absorption spectrum of curcumin.

These findings illustrate the diverse pH responsiveness of plant pigments, particularly anthocyanins, flavonoids, carotenoids, and betacyanins. The chemical transformations observed in anthocyanins and flavonoids highlight their potential as natural pH indicators, whereas the stability of carotenoid-based pigments underscores their resistance to environmental fluctuations in pH.

4. DISCUSSION

4.1. Interaction of Natural Pigments with pH

The color change of natural pigments in response to pH is caused by modifications in their molecular structure, which affect the conformation and stability of pigments under various conditions. Anthocyanins, flavonoid pigments found in *Clitoria ternatea* and *Impatiens balsamina*, undergo structural changes depending on pH. In acidic conditions (pH <3), anthocyanins appear red in the flavylium cation form. In contrast, at neutral to basic pH, they transition into the quinonoidal form, which exhibits purple or blue hues, before ultimately degrading at higher pH levels [47], [48]. Betalains, found in Beta vulgaris, are more stable within the pH range of 4–6 but undergo degradation in alkaline environments due to oxidation and hydrolysis of their indole ring, leading to a loss of red or yellow color in food applications [49]. In contrast, carotenoid pigments, such as those found in Tagetes erecta, exhibit high stability across different pH levels due to their non-polar nature, resulting in minimal color changes [50]–[52].

Curcumin, the primary phenolic pigment in turmeric (Curcuma longa), exhibits a more limited color response than anthocyanins. Under acidic to neutral conditions, curcumin retains its yellow color; however, at basic pH (7.5–8.5), its phenolic groups undergo deprotonation, causing a shift to a red-orange hue [53], [54]. Nevertheless, the pH-responsive range of curcumin is relatively narrow, and the pigment is also susceptible to photochemical degradation and oxidation under certain conditions, which may impact its stability in industrial applications. Based on these characteristics, it can be concluded that anthocyanins demonstrate high sensitivity to pH changes with prominent color shifts. At the same time, betalains remain stable under slightly acidic conditions, carotenoids exhibit stability across various pH levels, and curcumin shows limited color response within a narrow pH range. This understanding has significant implications in the food, pharmaceutical, and pH indicator technology industries.

4.2. Pigment Response

Tagetes erecta (*marigold*) contains lutein and zeaxanthin (*carotenoids*), which are chemically stable due to their conjugated polyene structure. A study by Fernández-García et al. [12] demonstrated that carotenoids remain stable across a pH range of 2–9, making them suitable for food or cosmetic coloring applications. This stability also supports their use in packaging materials exposed to variable environmental conditions. Similarly, Beta vulgaris (beet) extract contains betalains, which are prone to oxidation at high temperatures or under light exposure [55]. However, betalains can be preserved for textile or decorative paper coloring applications when incorporated into paper substrates with protective coatings (such as natural polymers).

Impatiens balsamina contains anthocyanins with free hydroxyl groups susceptible to pH. Research by Yoshida et al. [56] revealed that anthocyanins from this flower undergo a hyperchromic shift (increased color intensity) in alkaline conditions, making them ideal indicators. An example application is an indicator paper for detecting alkaline leaks in industrial settings. *Clitoria ternatea*, on the other hand, contains ternatin (a modified anthocyanin) that remains stable between pH 3–8. Kazuma et al. [57], the acylation structure of ternatin enhances its stability, allowing for applications in a wide pH range, such as environmental sensors or innovative packaging.

Curcumin exhibits a limited response to extreme pH conditions due to its low number of reactive functional groups. The molecule primarily contains phenolic hydroxyl groups and a diketone moiety, which contribute to its antioxidant properties but offer minimal responsiveness to drastic pH changes [53]. Under highly acidic (pH < 3) or alkaline (pH > 9) environments, curcumin undergoes degradation or structural rearrangements, leading to reduced stability and functionality [58]. This instability limits its direct application in systems requiring pH-dependent interactions.

The restricted but tunable reactivity of curcumin underscores its potential in niche applications. For example, its pH-dependent optical properties, when combined with nanomaterials like nanocellulose or metal-organic frameworks (MOFs), have been explored for real-time monitoring of food spoilage or environmental pollutants [59], [60]. These advancements align with broader efforts to leverage natural pigments in sustainable technologies, thereby bridging the gap between biochemical stability and industrial practicality.

4.3. Integration of Pigments into Paper Substrates

Integrating natural pigments into paper requires strategies to enhance color retention and stability. One approach involves the adsorption of pigments onto cellulose fibers, where negatively charged paper fibers can bind positively charged pigments, such as anthocyanins, in acidic conditions through electrostatic interactions [61]. Another method is encapsulation using biopolymers, where alginate or chitosan act as protective coatings to increase pigment stability against oxidation [62]. Cross-linking agents such as tannins or citric acid can reinforce pigment-fiber binding, improving durability and color fastness [63]. These techniques align with green chemistry trends that prioritize bio-based materials, as demonstrated by the successful development of pH indicator paper derived from red cabbage extract, which has been widely adopted in schools for chemistry education.

Beyond educational applications, natural pigment-infused paper has been explored in health diagnostics, innovative packaging, and environmental monitoring. In medical diagnostics, pH-sensitive paper has been developed for urine testing, particularly in detecting urinary tract infections (UTIs). Similarly, in food packaging, anthocyanin-based active packaging derived from grape skin has been employed to monitor fish freshness, with a visible color change from red (fresh) to blue (spoiled) [64].

5. CONCLUSION

This investigation underscores the significant potential of plant-derived pigments as pH-sensitive indicators in paper applications. While the study demonstrates their effectiveness in providing eco-friendly alternatives to synthetic dyes, further research is necessary. Future investigations should focus on the durability and stability of these pigments in practical settings, addressing challenges like time-dependent degradation and environmental interactions.

Optimizing extraction and paper-making processes will be essential to scale up production while ensuring consistent performance across batches. A standardized approach to sourcing and cultivating flowers, as well as the refinement of pigment incorporation techniques, will be critical for the successful development of these materials.

Additionally, analyzing the overall ecological impact of the paper-making process, including energy and water usage, is crucial to assessing the sustainability of using natural pigments in industrial applications. The scientific community should prioritize research into the broader applications of these pH-sensitive papers, particularly in health diagnostics and food packaging, ensuring that commercial barriers, such as production costs and market acceptance, are addressed.

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