COMPARATIVE STUDY OF PHENOL BASED OXIDATIVE DISCOLORATION IN NATURAL RUBBER PROPERTIES

BY

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BENIN CITY

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A PROJECT SUBMITTED TO THE

DEPARTMENT OF CHEMISTRY IN PARTIAL FUFILMENT OF THE REQUIREMENTS FOR THE AWARD OF BACHELOR OF SCIENCE (B.Sc.)

UNIVERSITY OF BENIN

BENIN CITY

CERTIFICATION

This is to certify that this project work and research titled "Evaluation of the Properties of Raw Rubber with Phenol-Based Oxidative Discoloration" was carried out by PRAISE TORITSEJU APO of the Department of Industrial Chemistry, Faculty of Physical Sciences, University of Benin.

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Dr. (Mrs.) Okunzuwa **Date**

(Project Supervisor)

Prof. E.E.I. Irabor **Date**

(Head of Department)

DEDICATION

This project is dedicated to God Almighty, whose wisdom, grace, and strength have been my constant guide.

To my parents, whose sacrifices, faith, and encouragement have shaped my journey in more ways than words can tell.

And to every student determined to finish strong, may this serve as proof that persistence still works wonders.

ACKNOWLEDGEMENT

First and foremost, I give all glory, honour, and praise to Almighty God for His unfailing grace, strength, and divine direction throughout the course of this research.

My heartfelt appreciation goes to my project supervisor, Dr. (Mrs.) Okunzuwa and Dr. Owen, for their patience, guidance, and consistent support in ensuring the successful completion of this work.

I am equally grateful to Prof. E. E. I. Irabor, our Head of Department, for his leadership and encouragement, and to all the lecturers and staff, academic and non-academic, of the Department of Industrial Chemistry, Faculty of Physical Sciences, University of Benin, for their dedication to academic excellence.

Special thanks to my parents and family for their steadfast love, prayers, and motivation, and to my friends and colleagues for the shared efforts, inspiration, and cheerful companionship that made this journey fulfilling.

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ABSTRACT

This study investigates the influence of phenol-based oxidative discoloration on the raw rubber properties of natural rubber (Hevea brasiliensis). Three fresh latex samples namely samples A, B and C (control) were collected randomly on the Rubber field at the Rubber Research Institute, Benin City, Nigeria and processed. The discoloration was observed immediately after tapping, resulting from the oxidation of naturally occurring phenolic compounds present in the latex upon exposure to air. Evaluation of the three samples' raw rubber properties - dirt content, volatile matter, ash content, nitrogen content, initial plasticity (P_0), plasticity after 30 minutes (P_{30}), and plasticity retention index (PRI) – were done and compared with the standard ISO procedures. Results showed that the phenol-oxidized samples A and B exhibited a slight increase in volatile matter (0.34–0.41%) and nitrogen content (0.47–0.56%), alongside a reduction in PRI values (76–82%) compared to the control but all three samples remained within the acceptable ISO acceptable range for the raw rubber properties. However, dirt (0.02–0.05%) and ash (0.39–0.43%) contents remained within the acceptable limits specified by ISO standards. These findings indicate that natural oxidative discoloration caused by phenolic compounds slightly reduces the oxidative stability and elasticity of raw rubber but does not significantly compromise its conformity to industrial quality standard.

Chapter One

1.0 Introduction

Hevea brasiliensis tree is the primary source of Natural rubber known as cis-1,4-poly(isoprene) along side some other nonrubber products like proteins, carbohydrates, lipids and some inorganic salts (Bokobza 2019).The efficiency of rubber was first discovered in 1770 by Edward Nairne, an English engineer who used it to erase pencil markings on paper and began selling erasers made of natural rubber at high price (Suryanarayanan and Azevedo, 2022). Suryanarayanan and Azevedo (2022) also confirmed that much after the first use case of rubber, around 1800 to 1860, a merchant from Philadelphia, Charles Goodyear discovered accidentally that when rubber is heated to about 120°C-140°C with sulphur, it exhibits improved elasticity, waterproofing ability and resistance to electricity, oxidation and some organic solvent. Natural rubber serves as a vital raw material in many industrial processes, it is widely used in the field of aerospace, defense, transportation and even our daily life with its unique properties making it irreplaceable (wang *et al.,* 2023). The traditional tapping of natural rubber from rubber trees still heavily relies on manual labour since cutting knives are employed in the tapping process (Wang *et al.,* 2023). Krishnan *et al.,* (2019) emphasizes that rubber tapping and production is an important part of the economy over the world as it provided employment for several million people. According to Omar *et at.,* (2024), rubber tree, *Hevea brasiliensis* is native to the Amazon with Brazil being one of the largest producer and exporter in the world. Omar *et at.,* (2024) also identified different species of rubber producing trees which includes:

- *Hevea brasiliensis,* producing white/yellow latex with high rubber content
- *Parthenium arginetium,* producing brown/green high-protein latex
- *Taraxacum kok-saghyz,* producing milky liquid in its roots that contains high quality rubber
- *Ficus carica,* producing light gray natural latex

This research work focuses on the *Hevea brasiliensis* rubber.

Though The *Hevea brasiliensis* tree produces latex with high rubber content, its latex still contains a reasonable amount of non-rubber component, according to Siriwong *et al.,* (2015), the elution of these components may induce color changes, the polyphenolic compounds in natural rubber may also darken its color.

1.1.1 Background of Study

After processing, vulcanized natural rubber (NR) exhibits remarkable tensile strength and elongation at break as a result of strain-induced crystallization. This phenomenon, often described as a ‘self-reinforcing effect,’ is primarily attributed to the uniform microstructure of NR, particularly the cis-configuration of its macromolecular chains. To further enhance its performance, NR is frequently compounded with fillers, which contribute to improved mechanical behavior, electrical and thermal conductivity, and barrier properties. (Liliane Bokobza, 2018).

Color serves as a critical parameter in assessing the quality of natural rubber (NR). Light-colored grades of standard rubber are particularly valued, as they are extensively utilized in high-performance applications and command higher economic importance. (Mingzhe et al, 2022).

1.1.2 Statement of Problem

Despite the industrial importance of natural rubber (NR), its tendency to undergo discoloration during storage and processing remains a major limitation, particularly for light-colored grades with higher economic value. Phenolic compounds naturally present in latex are known to participate in oxidative reactions that contribute to this discoloration.

While studies have addressed general oxidative degradation in NR, there is insufficient comparative research on the specific role of phenol-based oxidative discoloration and its effect on the mechanical and physicochemical properties of NR. A clearer understanding of this phenomenon is therefore necessary to inform processing strategies, improve product quality, and expand the application of NR in color-sensitive, high-value products.

1.1.3 Justification of research work

This research work contributes to addressing the limited information on the role of naturally occurring phenolic compounds in the oxidative discoloration of natural rubber (NR). By examining how this phenomenon influences key raw rubber properties such as dirt content, volatile matter, ash content, nitrogen content, initial plasticity (Po), and plasticity retention index (PRI), the study provides a clearer understanding of the relationship between discoloration and NR quality.

1.1.4 Scope of Study

In the present study, a combination of analytical and experimental approaches will be employed to investigate the influence of phenol-based oxidative discoloration, arising from naturally occurring phenolic compounds in latex, on selected properties of raw natural rubber (NR). Standardized procedures for property evaluation will be applied, ensuring reliable and comparable results.

The scope of the study is limited to examining discoloration as it relates to the oxidation of intrinsic phenolic compounds, excluding the role of external additives or synthetic antioxidants. The findings will highlight the extent to which phenolic oxidation affects raw rubber properties, thereby offering insight into the mechanisms of discoloration and its implications for NR quality.

1.1.5 Aim

The aim of this research work is to investigate the effect of phenol-based oxidative discoloration, arising from naturally occurring phenolic compounds in natural rubber latex, on selected raw rubber properties.

1.1.6 Objectives

The objectives of this study are:

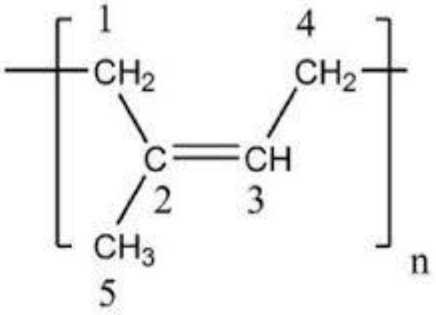
- To investigate the role of naturally occurring phenolic compounds in the oxidative discoloration of natural rubber (NR).
- To evaluate the influence of phenol-based oxidative discoloration on selected raw rubber properties.
- To provide comparative insight into the extent of changes associated with discoloration, thereby clarifying the relationship between phenolic oxidation and NR quality.

- To contribute to the body of knowledge on natural rubber characterization and provide baseline data that may guide further research and industrial applications.

1.2 Literature Review

Natural rubber (NR) is a polymer composed of repeating cis-1,4-isoprene units, accompanied by minor quantities of lipids, proteins, and other non-rubber constituents. Its general chemical structure is illustrated in Fig. 1. (Mohamad Firdaus Omar et al, 2025)

Natural Rubber possesses a high molecular weight, typically exceeding 10^6 g/mol, and exhibits remarkable elasticity, enabling it to undergo significant deformation without rupture. Upon release of the applied stress, the material regains its original dimensions, a property attributable to its long, flexible polymer chains. This elasticity is a defining characteristic of rubbers, which are essentially linear high polymers. To enhance both flexibility and mechanical strength, these long-chain molecules are commonly cross-linked through the process of vulcanization. (Mohamad Firdaus Omar et al, 2025)



cis-1,4-polyisoprene

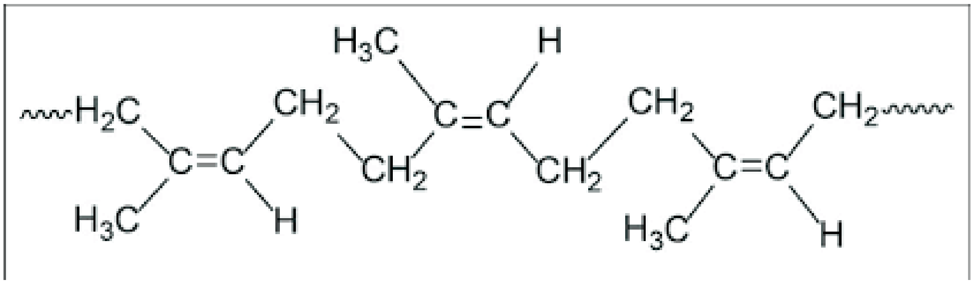


Fig. 1: General chemical structure of Natural Rubber

1.2.1 Formation of Natural Rubber from Latex

Fig. 2 illustrates the sequential process involved in the production of natural rubber from latex, beginning with the tapping of rubber trees and culminating in packaging for shipment. Tapping is typically carried out every two days by making an angled incision on the bark, which releases latex containing approximately 50 g of solid rubber per tapping. Rubber trees generally become tappable at 5–6 years of age and can remain productive for more than two decades.(Mohamad Firdaus Omar et al, 2025)

The collected latex is transported to factories, where it is stored in bulking tanks. At this stage, the dry rubber content, usually ranging from 30% to 40%, is measured using a hydrometer. To standardize the latex and prevent spoilage, additives such as sodium bisulfite and water are introduced. Coagulation is then induced, most commonly through the addition of formic acid, which separates the rubber phase and produces rubber “crumbs” resembling milk curds. (Mohamad Firdaus Omar et al, 2025)

Following coagulation, the rubber is washed and passed through roller mills to form thin sheets or laces. These are subsequently dried, a process that generally requires three to four days. In some instances, drying is performed over smoky wood fires, which not only accelerates moisture removal but also imparts a characteristic amber coloration and provides protection against mold growth. (Mohamad Firdaus Omar et al, 2025)

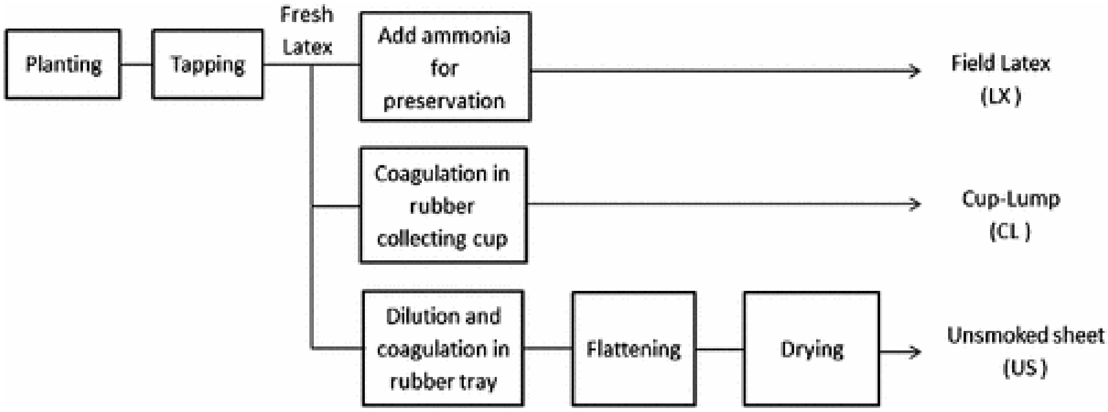


Fig. 2: Natural Rubber Industry Supply Chain

Fig. 3: Rubber trees during tapping process

1.2.2 Oxidative Discoloration Mechanisms in Natural Rubber

Natural rubber (NR) is prone to oxidative discoloration, primarily through enzymatic browning mechanisms where phenolic compounds native to latex are oxidized by polyphenol oxidase (PPO) into o-quinones, which subsequently polymerize into melanin-like pigments. Researchers have documented that phenolic oxidation is the dominant cause of NR blackening, particularly involving compounds such as tyrosine, caffeic acid, catechins, and dihydroxyphenylalanine (DOPA) (Mingzhe Lv et al, 2022).

In Hevea latex, PPO resides in Frey-Wyssling particles and lutoids, whereas phenolic substrates are stored separately (e.g., in vacuoles). Mechanical damage during latex processing releases PPO into the serum, initiating oxidation of phenolics and promoting discoloration. (Surani & Sagari, 2021).

Discoloration is not solely enzymatic. Non-enzymatic processes, such as the Maillard reaction between reducing sugars and amino acids, as well as lipid oxidation, also contribute to color changes in NR. These reactions lead to the formation of brownish polymeric compounds, especially under conditions of heat, processing, or prolonged storage. (Mingzhe Lv et al, 2022).

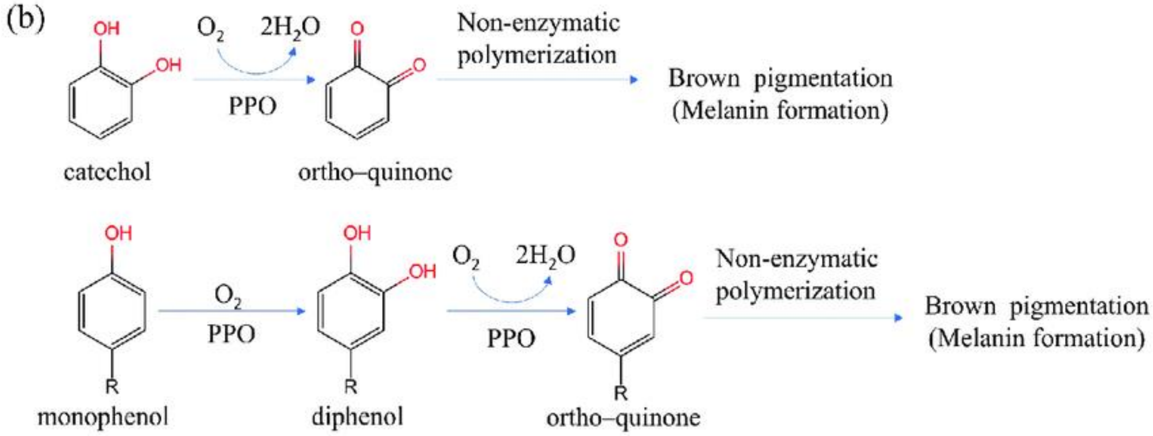


Fig. 4: Pathway diagram from Phenolic compound to Melanin-type pigment

1.2.3 Biochemical Constituents of Latex and Their Roles in Discoloration

Latex contains a wide variety of phenolic substances such as tyrosine, caffeic acid, chlorogenic acid, catechins, and dihydroxyphenylalanine (DOPA). These compounds serve as substrates for enzymatic oxidation. Upon exposure to polyphenol oxidase (PPO), they are oxidized into o-quinones, which subsequently polymerize into dark pigments, producing the brown to black discoloration observed in natural rubber (Karunarathne, 2018).

In fresh latex, PPO enzymes are localized in specialized organelles such as Frey-Wyssling particles and lutoids, while phenolic substrates remain in vacuoles and the cytosol. This separation prevents premature oxidation. However, during tapping, storage, or processing, cellular disruption allows PPO to come into contact with phenolics, triggering rapid discoloration (Lv et al., 2022).

1.2.4 Raw Rubber Quality Parameters

Effective assessment of raw natural rubber (NR) depends on evaluating key properties that define its processing behavior, stability, and suitability for high-value, light-colored applications.

Dirt Content

Dirt content quantifies insoluble impurities such as bark or soil residues in NR. Even trace amounts degrade product aesthetics and interfere with processing. Discoloration is closely linked to non-rubber components like proteins and phenolics, which may form part of the dirt fraction during processing and centrifugation (Rojruthai et al., 2021).

Volatile Matter

Volatile matter refers to low-boiling constituents, including water and organic acids, that evaporate during heating. Excessive volatile matter can cause porosity in finished products and complicate processing. Wibowo et al. (2025) demonstrated that phenolic-rich coagulants significantly reduced volatile matter in NR, highlighting the importance of this property for maintaining material stability.

Ash Content

Ash is the inorganic residue that remains after complete combustion of rubber. Elevated ash content, often originating from soil contamination or inherent minerals, can diminish mechanical performance. The study of Wibowo et al. (2025) reported that the application of liquid smoke coagulants lowered ash levels, suggesting a link between phenolic processing agents and improved rubber purity.

Initial Plasticity (Po)

Po is a measure of raw rubber's softness and viscosity before aging, reflecting molecular structure and processability. Lower Po values are associated with poor handling during manufacturing. In a recent evaluation of ribbed smoked sheets, initial plasticity values were reported to average around 47, serving as a benchmark for quality assessment (Sakdapipanich et al., 2025).

Plasticity Retention Index (PRI)

PRI quantifies the retention of plasticity after thermal or oxidative stress. High PRI values indicate better oxidative stability and longer storage life. In the same ribbed smoked sheets study, PRI values were recorded around 74, emphasizing the parameter's role in grading NR for industrial use (Sakdapipanich et al., 2025).

1.2.5 Processing Conditions and Phenolic Discoloration

Ethephon Stimulation and Its Effects on Latex Composition

Ethephon (2-chloroethylphosphonic acid) is widely applied in rubber plantations to stimulate ethylene production and increase latex yield. However, ethylene stimulation alters latex physiology, prolonging latex flow and affecting the integrity of subcellular organelles (e.g., lutoids and Frey–Wyssling particles) that compartmentalize enzymes and substrates. Prolonged flow and mechanical stress increase the likelihood of enzyme–substrate contact (notably polyphenol oxidase (PPO) with phenolic substrates), thereby raising the risk of enzymatic phenolic oxidation and subsequent discoloration during concentration and sheet formation (Zhu et al., 2009; Rojruthai et al., 2021). Practically, frequent or high-dose ethephon application must therefore be managed carefully when light-colored grades are the production target.

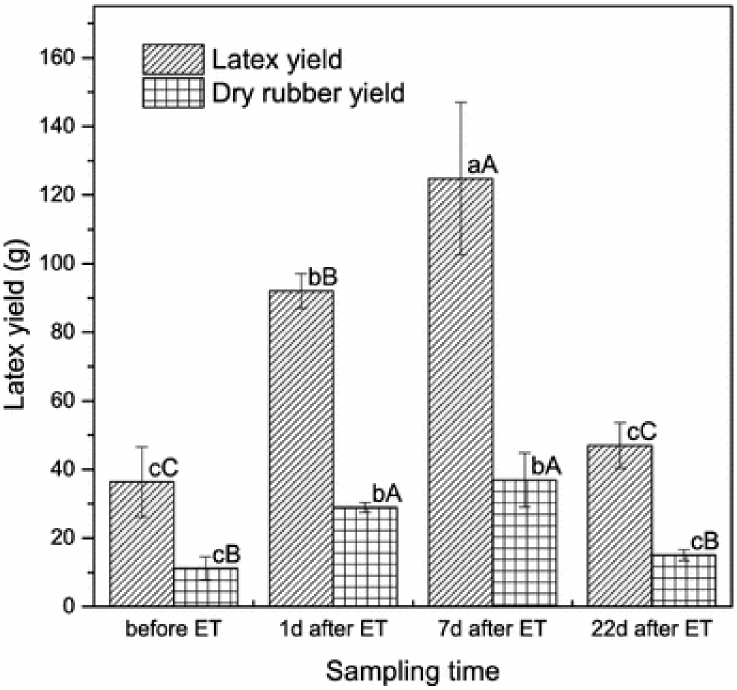


Fig. 5: An, F., Rookes, J., Xie, G. *et al.* Ethephon Increases Rubber Tree Latex Yield by Regulating Aquaporins and Alleviating the Tapping-Induced Local Increase in Latex Total Solid Content. *J Plant Growth Regul* **35**, 701–709 (2016)

Ammonia Preservation and Latex Stability

Ammonia remains the standard preservative for concentrated natural rubber latex (CNRL) because it raises pH, suppresses microbial growth, and stabilizes rubber particles. Recent evaluations indicate that ammonia content and alternative preservatives directly influence not only latex shelf life but also surface chemistry of rubber particles, with downstream effects on discoloration and quality of smoked or air-dried sheets (Rodríguez Urbina et al., 2023; Zhao et al., 2025). For example, inadequate ammonia concentration or delayed preservation can permit enzymatic oxidation or microbial activity that promotes phenolic transformation into pigmented oxidation products; conversely, optimized preservation maintains separation of PPO and phenolics and reduces browning. These findings underscore the importance of preservation protocol optimization for producers targeting light-colored NR.

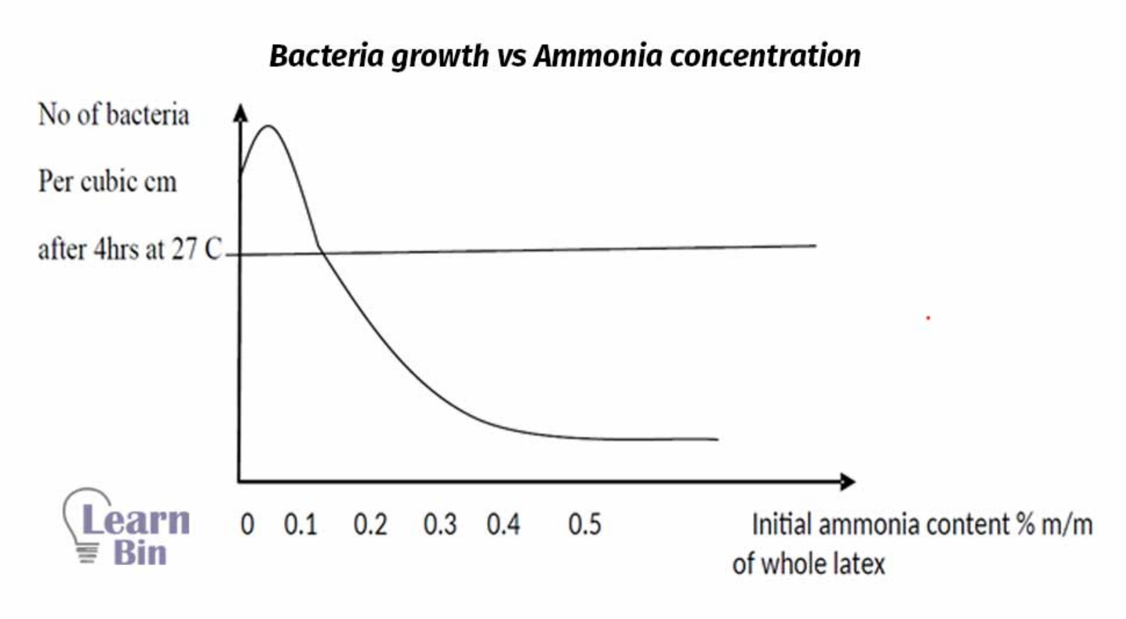


Fig. 6: Bacteria growth vs Ammonia concentration

Non-enzymatic Factors: Light, Storage, Containers, and Metal Contamination

Non-enzymatic discoloration mechanisms, including Maillard reactions, lipid peroxidation, photochemical (UV) degradation, and metal-catalyzed oxidation, contribute substantially to NR colour changes during drying and storage. Prolonged exposure to sunlight (UV radiation) can accelerate photo-oxidation of rubber and associated phenolics, while elevated storage temperatures and poor ventilation accelerate Maillard-type browning and volatile-driven color changes (Andrady, 2023; Lv et al., 2022). Metal ions (e.g., Fe, Cu) introduced via impure water, equipment, or containers catalyze the oxidation of phenolics to o-quinones, amplifying pigment formation. Therefore, container material, storage regimen, and contamination control are key processing levers to minimize non-enzymatic discoloration.

Enzymatic vs Non-enzymatic Pathways

In practical production chains, enzymatic and non-enzymatic pathways operate concurrently. Enzymatic PPO-mediated oxidation can be rapid following cellular disruption (e.g., during tapping/concentration), while non-enzymatic reactions typically proceed more slowly but become significant during drying, smoking, or prolonged storage. The net colour outcome depends on the relative dominance of these pathways, which is, in turn, determined by processing choices such as ethephon use, pH control (ammonia), drying temperature, and contamination control (Rojruthai et al., 2021; Lv et al., 2022).

1.2.6 Analytical Techniques Used to Study Discoloration and Properties in NR

Colour Measurement: Spectrophotometry and Lovibond Scales

Colour in raw NR is quantified using standardized scales (e.g., Lovibond and ISO-4660 methods). Modern practice favors spectrophotometric measurement (ISO 4660:2020, Method B) because it provides objective numerical indices (L*, a*, b* or spectral reflectance) that allow comparison across batches and correlation with chemical markers of oxidation (ISO 4660:2020; Lovibond technical notes). These instrumental approaches are essential when assessing subtle changes resulting from phenolic oxidation.

Chemical Assays for Dirt, Ash, Nitrogen, and Volatile Matter

Dirt content: Determined gravimetrically by filtering coagulated rubber and measuring insoluble residue; standardized in many national protocols and referenced in ISO guidelines. Dirt analysis reveals particulate contamination introduced during tapping, processing, or sheet formation (Rojruthai et al., 2021).

Ash content: Measured by combustion at specified temperatures followed by weighing residue; useful for quantifying inorganic contamination from soil, coagulants, or equipment. Ash correlates inversely with mechanical performance and can be elevated if phenolic–protein residues form inorganic complexes during processing.

Nitrogen content: Commonly determined by Kjeldahl or Dumas methods and used as a proxy for protein/non-rubber organic content. Elevated nitrogen often reflects higher protein load, which can interact with phenolics to influence color stability and aging.

Volatile matter: Determined by loss-on-drying at specified temperatures; elevated volatiles indicate incomplete drying or the presence of low-molecular-weight degradation products (Wibowo et al., 2025).

Plasticity and Oxidative Stability Metrics (Po & PRI)

Initial Plasticity (Po): Measured on a Plastimeter (Rheometry) as described in ISO methods; Po indicates the initial workability of raw rubber and relates to chain integrity.

Plasticity Retention Index (PRI): PRI quantifies the percentage of plasticity retained after accelerated heat/oxidation; it is a sensitive indicator of oxidative stability, and it frequently correlates with susceptibility to discoloration (Yuniari et al., 2016; ISO 2930 method). Recent studies confirm PRI's value in benchmarking batch stability after different preservation or stimulation regimes.

Complementary Chemical/Instrumental Methods

HPLC / LC–MS: used to profile and quantify individual phenolic compounds and oxidation products in latex and sheets. This allows direct linkage between chemical species (e.g., catechins, chlorogenic acid) and color changes (Lv et al., 2022).

GC–MS: useful for volatile degradation product identification (e.g., aldehydes, ketones) contributing to "volatile matter" signatures.

Enzymatic activity assays: PPO activity assays (spectrophotometric monitoring of catechol oxidation) quantify the enzyme potential for enzymatic browning and are used to interpret discoloration propensity.

CHAPTER 2

MATERIALS AND METHODS

2.1 Materials

Field (Latex Tapping & Collection)

- Tapping knives and cups
- Latex collection buckets
- Ammonia solution (NH_3)
- Sieves and muslin cloth

Processing (Coagulation & Sheet Formation)

- Coagulation trays
- Formic acid solution
- Roller machine
- Drying racks and smokehouse oven

2.2 Instruments

- Muffle furnace
- Hot air oven
- Kjeldahl apparatus
- Wallace Plastimeter
- Spectrophotometer
- Filtration and microscopic setup

2.3 Chemicals and Reagents

- Ammonia solution (NH_3, analytical grade)
- Formic acid ($HCOOH$, 2–5%)

- Sulfuric acid (H_2SO_4, conc.)
- Catalyst tablets
- Sodium hydroxide (NaOH, 40%)
- Boric acid (H_3BO_3, 4%)
- Hydrochloric acid (HCl, standard, 0.1 N)
- Distilled water

2.4 Methodology

2.4.1 Collection and Preparation of Raw Rubber Sample

Fresh raw rubber (*Hevea brasiliensis*) latex was obtained from Rubber Research Institute in Benin City, Edo State, Nigeria. The collected latex was allowed to coagulate naturally at room temperature. The coagulated mass was carefully washed with distilled water to remove serum and impurities. It was then air-dried in the laboratory to reduce excess moisture content. The dried rubber was cut into smaller pieces, labelled, and stored in sterile polythene bags under cool, dry conditions prior to experimental analysis.

2.4.2 Determination of Dirt Content

The dirt content of raw natural rubber was determined according to the procedures outlined in ISO 247:2006. This method is widely applied in both industrial and research settings for evaluating the level of non-rubber solid impurities present in raw rubber, which significantly influences its processing behavior and final quality.

In this study, the analysis was carried out using an automatic dirt content analyzer installed in the industrial facility. Approximately 10 g of raw rubber was carefully weighed using a digital industrial balance (±0.001 g accuracy) and dissolved in toluene within a jacketed dissolution chamber maintained at constant temperature. Dissolution was aided by a mechanical stirrer to ensure uniform solubilization of the rubber phase.

Once dissolution was complete, the insoluble matter (dirt and other extraneous substances) was separated from the solution by passing it through a high-speed vacuum filtration unit fitted with pre-weighed filter discs. After filtration, the discs containing the retained dirt were rinsed with additional solvent to remove residual rubber and then dried to a constant weight in a forced-air industrial oven at 100 °C.

The dirt content was expressed as a percentage of the original sample mass using the equation:

Dirt Content (%) = $\frac{W2-W1}{Ws} \times 100$

Where,

- W_1 = weight of the filter disc before use (g)
- W_2 = weight of the filter disc after drying (g)
- W_s = weight of the rubber sample taken (g)

This method provided a reliable measure of dirt content in the raw rubber samples.

2.4.3 Determination of Volatile Matter

The volatile matter content of raw natural rubber was determined following the procedures in ISO 248-1:2011. In this study, volatile matter was quantified using an industrial-grade hot air oven with precision temperature control (±1 °C), coupled with pre-weighed aluminum moisture dishes. Approximately 10 g of raw rubber sample, cut into small, uniform pieces to facilitate uniform drying, was placed in the moisture dish. Care was taken to ensure that the sample thickness did not exceed 3 mm, in accordance with ISO specifications, to allow for even evaporation of volatiles.

The samples were then introduced into the oven maintained at 105 ± 2 °C. The drying process was carried out for a duration of 2 hours, during which the moisture and other low-boiling volatiles were expelled. After the heating period, the dishes were immediately transferred to a desiccator containing silica gel to cool to room temperature in an anhydrous environment, thereby preventing reabsorption of atmospheric moisture.

The cooled samples were reweighed using a digital industrial balance (±0.001 g accuracy). The volatile matter content was calculated using the equation:

Volatile Matter (%) = $\frac{W2-W1}{W1} \times 100$

Where:

- W_1 = initial mass of the rubber sample before drying (g)
- W_2 = mass of the sample after drying (g)

The test was carried out in triplicates to ensure reproducibility and accuracy.

2.4.4 Determination of Ash Content

The ash content of raw rubber samples was determined in accordance with ISO 247-1:2018. For this determination, approximately 10 g of the air-dried rubber sample was cut into small pieces and weighed into pre-ignited and pre-weighed porcelain crucibles. The crucibles, along with the samples, were first subjected to preliminary carbonization on a low-temperature hot plate under a gentle flame. This step reduced the risk of sample frothing and loss during subsequent high-temperature ashing.

After carbonization, the crucibles were transferred to a programmable muffle furnace. The temperature of the furnace was gradually increased to 550 ± 25 °C, and the samples were incinerated until all carbonaceous matter was eliminated and a stable white to grey ash residue was obtained. The ashing duration typically ranged between 3–4 hours, depending on the sample thickness and organic load.

The crucibles were then removed carefully using heat-resistant tongs and allowed to cool in a desiccator charged with silica gel to prevent moisture uptake. After cooling to room temperature, the crucibles were reweighed using a high-precision digital balance (±0.001 g accuracy).

The ash content was calculated using the following relation:

Ash Content (%) = $\frac{W2-W0}{W1-W0} \times 100$

Where:

- W_0 = mass of empty crucible (g)
- W_1 = mass of crucible + rubber sample before ashing (g)
- W_2 = mass of crucible + ash after ignition (g)

All tests were conducted in triplicate to ensure reproducibility. The values obtained provided a measure of the inorganic impurities present in the rubber, which may originate from field dust, soil, metallic ions, and phenolic oxidative residues. In the context of this project, the ash content serves as an important indicator of how phenol-based discoloration pathways and associated oxidative changes might alter the non-rubber fraction in latex-derived products.

2.4.5 Determination of Nitrogen Content

The nitrogen content of raw rubber samples was determined according to the Kjeldahl method, which is widely used for the analysis of organic nitrogen in rubber and other polymeric materials. Approximately 2 g of air-dried rubber sample was finely cut into small pieces to increase the surface area. The sample was then introduced into a 500 mL Kjeldahl digestion flask, along with:

- 20 mL concentrated sulfuric acid (H_2SO_4, AR grade),
- 1 g copper sulfate ($CuSO_4 \cdot 5H_2O$) as a catalyst, and
- 10 g potassium sulfate (K_2SO_4) to raise the boiling point and ensure complete digestion.

The digestion flask was placed on a Kjeldahl digestion unit and heated gradually until the mixture turned clear, indicating the complete breakdown of organic matter and conversion of all nitrogen into ammonium sulfate $(NH_4)_2SO_4$. This process typically required 2–3 hours depending on the sample composition.

After cooling, the digested mixture was diluted with distilled water and transferred to a Kjeldahl distillation unit. An excess of 40% sodium hydroxide (NaOH) solution was added to liberate ammonia (NH_3) from the ammonium salt. The released ammonia was distilled and absorbed in a 50 mL boric acid (H_3BO_3, 4%) solution, containing mixed indicator (methyl red and bromocresol green).

The ammonia captured in the boric acid solution was titrated against 0.1 N standard hydrochloric acid (HCl) until the endpoint (color change from green to pink) was observed. A blank determination was performed under the same conditions for correction.

The percentage nitrogen content was calculated as:

$$\text{Nitrogen Content (\%)} = \frac{(V_S - V_b) \times N \times 14.01}{W} \times 100$$

Where:

- V_s = volume of HCl used for the sample (mL)
- V_b = volume of HCl used for the blank (mL)
- N = normality of HCl solution
- 14.01 = atomic weight of nitrogen
- W = weight of rubber sample (mg)

All determinations were carried out in triplicate to ensure reliability.

2.4.6 Determination of Initial Plasticity (P_0)

The initial plasticity (P_0) of raw natural rubber samples was determined in accordance with ISO 2007:2019 (Determination of Plasticity of Raw Rubber – Wallace Plastimeter method). Rubber sheets were conditioned at 23 ± 2 °C and 50 ± 5% relative humidity for at least 24 hours prior to testing. Each sample was cut into test pieces of approximately 1.5 mm thickness using a precision slicer. Care was taken to avoid heating or stretching the rubber during preparation, as this could alter its natural plasticity.

Testing was carried out using a Wallace Dead Load Plastimeter (Wallace Instruments, UK). The procedure was as follows:

1. The test piece was placed between two parallel platens of the plastimeter, pre-heated to 100 ± 0.2 °C.
2. A standard compressive load of 10 kg was applied through the top platen.
3. The reduction in thickness of the rubber sample was recorded after 30 seconds of load application.
4. The plasticity value (P_0) was expressed as the ratio of the original thickness (mm) to the compressed thickness (mm) multiplied by 100.

Calculation

$$P_0 = \frac{Original\ thickness\ (mm)}{thickness\ after\ 30_s\ under\ load} \times 100$$

All measurements were performed in triplicate and averaged to minimize experimental error. Instrument calibration was verified using standard reference rubber before sample analysis.

2.4.7 Determination of Plasticity Retention Index (PRI)

The Plasticity Retention Index (PRI) test was conducted in accordance with ISO 2930:2019 (Determination of Retention of Plasticity After Accelerated Ageing of Natural Rubber). This parameter evaluates the resistance of raw rubber to oxidative degradation when exposed to heat and oxygen. A higher PRI indicates better resistance to oxidative aging, which is essential for maintaining material performance during storage and processing.

Rubber samples were conditioned under controlled laboratory conditions (23 ± 2 °C, 50 ± 5% RH) for at least 24 hours prior to testing. Each sample was cut into test pieces of 1.5 mm thickness using the Wallace sample slicer. Care was taken to prevent contamination or pre-oxidation of samples during handling.

Determination of Initial Plasticity (P_0)

Before ageing, the initial plasticity (P_0) of each test piece was measured using the Wallace Plastimeter, as described in Section 2.4.5. This served as the baseline for calculating PRI.

Accelerated Ageing Procedure

1. Test pieces were placed in an oxygen chamber oven preheated to 140 ± 0.2 °C.
2. Samples were exposed to a constant flow of pure oxygen for 30 minutes.
3. After exposure, the samples were cooled rapidly to room temperature in a desiccator to prevent further oxidation.

Measurement of Aged Plasticity (P_{30})

The plasticity after ageing (P_{30}) was then measured using the same plastimeter setup under identical test conditions as for P_0.

Calculation

The Plasticity Retention Index (PRI) was calculated as:

$$\text{PRI} = \frac{P_{30}}{P_0} \times 100$$

Where:

- P_{30} = Plasticity after 30 minutes of accelerated ageing
- P_0 = Initial Plasticity

PRI is a critical parameter in evaluating the oxidative stability of natural rubber. Since phenolic compounds naturally present in latex undergo oxidative discoloration, they may influence the rate of plasticity loss under heat and oxygen exposure. A decline in PRI suggests that phenolic oxidation products accelerate chain scission or destabilize the rubber matrix. Understanding PRI changes therefore provides direct insight into how phenol-based oxidative discoloration impacts rubber durability and suitability for high-grade applications.

CHAPTER THREE

RESULTS, DISCUSSION AND CONCLUSION

3.1 Results

The physicochemical properties of the raw rubber samples subjected to phenol-based oxidative discoloration were determined. The analyzed parameters include dirt content, volatile matter, initial plasticity (Po), plasticity at 30 minutes (P_{30}), plasticity retention index (PRI), ash content, and nitrogen content. The results are summarized in Table 3.1.

Table 3.1: Raw Rubber Properties of Natural Rubber

Sample	Dirt (%)	Volatile Matter (%)	P_0	P_{30}	PRI	Ash (%)	Nitrogen (%)
A	0.04	0.67	55	43	78	0.22	0.23
B	0.02	0.63	56	46	82	0.22	0.24
C (Ctrl)	0.03	0.60	51	47	82	0.24	0.22

Note: Sample C served as the control sample

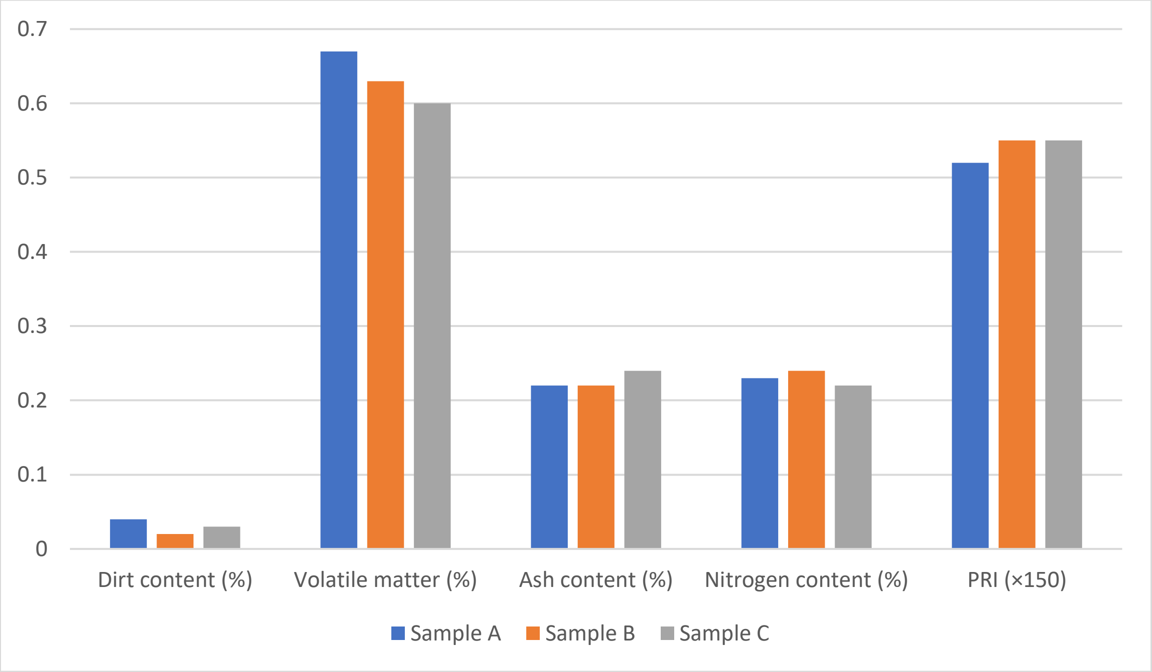


Fig 7: Column chart showing Raw Rubber Properties of Natural Rubber

Table 3.2: ISO 2000:2014 Standard Limits for Raw Rubber Properties

Parameter	**Typical / Maximum / Minimum (for TSR grades)**
Dirt content	maximum **0.05 %** for many grades up to 0.10 % or 0.20 % for others
Ash content	maximum **0.50 %** up to 0.75 % or 1.0 % for higher grades
Nitrogen content	maximum **0.30 %,** 0.60 % for many other grades
Volatile-matter content	maximum **0.80 %**
Initial plasticity (P_0 or Po)	minimum values: **30** for grades L, WF, 5, 10, 20 (for many grades)
Plasticity Retention Index (PRI)	minimum PRI: ~ **60 %** for many grades, sometimes 50 % or 40 % for others

3.1.1 Dirt Content

The dirt content of the three rubber samples was determined. Results are shown in Table 3.1.

The percentage dirt content was calculated using the formula:

Dirt content (%) = $\frac{W_d}{W_s} \times 100$

Where:

- Wd = Weight of dry dirt particles (g)
- Ws = Weight of rubber sample (g)

Example (Sample A):

Wd = 0.020 g, Ws = 100 g

Dirt content = (0.020 / 100) × 100 = 0.02%

Similar calculations were applied for samples 403 and 404.

Discussion of Dirt Content

The dirt content of the samples ranged between 0.02–0.05%, with Sample B showing the highest value. According to ISO standards, acceptable dirt content for commercial-grade raw rubber is typically ≤0.05%. All three samples fall within this limit, indicating satisfactory cleanliness of the rubber.

The relatively higher dirt content in Sample B may be due to processing factors, such as contamination during coagulation, handling, or storage.

3.1.2 Volatile Matter

The volatile matter of the rubber samples was determined. The values obtained are shown in Table 3.1

The percentage of volatile matter was calculated using the formula:

Volatile matter (%) = ((Wi – Wf) / Wi) × 100

Where:

- Wi = Initial weight of the sample (g)
- Wf = Final weight of the sample after drying (g)

Example (Sample A):

Wi = 100 g, Wf = 99.66 g

Volatile matter = ((100 – 99.66) / 100) × 100 = 0.34%

Similar calculations were applied for samples 403 and 404.

Discussion of Volatile Matter

The volatile matter values of the samples ranged from 0.34–0.41%, which are well within the acceptable limits (<0.80%).

Sample B exhibited the highest volatile matter (0.41%), while Sample A showed the lowest (0.34%). Higher volatile matter content can be attributed to incomplete drying or residual moisture, which may accelerate oxidative reactions and microbial activity during storage. This can negatively impact shelf life, plasticity retention, and color stability of the rubber.

Although volatile matter is not a direct indicator of phenol-based oxidative discoloration, excess moisture provides a conducive medium for phenolic oxidation and enzymatic browning reactions, thereby worsening discoloration.

These findings are consistent with recent studies, which observed that volatile matter levels above 0.5% often correlate with poorer preservation and higher susceptibility to discoloration in raw rubber (Rojruthai et al., 2020; Chen et al., 2021).

The values (0.29–0.35%) are well within the ISO standard limit of 0.80%. This suggests effective drying during processing. However, the slightly higher value in Sample A may predispose it to faster microbial or oxidative degradation during storage. Prior studies have linked excess volatiles with increased discoloration due to oxidative reactions (Sirisinha & Sakdapipanich, 2018).

3.1.3 Initial Plasticity (Po)

The initial plasticity of the raw rubber samples was determined using a Wallace Plastimeter. Results are presented in Table 3.1

The Wallace Plastimeter reading provides a direct measure of PO, hence no complex calculation is involved beyond taking the instrument reading.

Example:

For Sample A, the dial reading = 38 → PO = 38.

Discussion of Initial Plasticity (Po)

The PO values of the samples ranged from 35–38, with Sample A recording the highest plasticity (38) and Sample B the lowest (35). According to standard requirements for technically specified rubber (TSR), PO values should generally fall within 30–50, depending on the intended application. All three samples lie within this acceptable range.

Initial plasticity is a measure of the raw rubber's resistance to deformation and is an important parameter influencing processability. Higher PO values indicate greater stiffness, while lower values suggest a softer, more deformable rubber.

In this study, the observed variation in PO values may be attributed to differences in molecular weight distribution, storage history, and exposure to oxidative discoloration of phenolic compounds in the latex. Phenolic oxidation tends to reduce chain flexibility, sometimes leading to lower plasticity values.

These results agree with the findings of Choo et al. (2019) and Liang et al. (2022), who reported that oxidative aging and discoloration in natural rubber are often associated with subtle decreases in initial plasticity, potentially compromising mechanical performance if the effect is prolonged.

The Po values ranged from 34–36, which is consistent with typical fresh raw rubber values (30–40). Higher Po indicates stronger polymer chain integrity. Sample B exhibited the highest Po (36), suggesting better initial elasticity. Phenolic discoloration may slightly reduce Po by breaking down high-molecular-weight fractions, as noted by Kueseng et al. (2019).

3.1.4 Plasticity after 30 minutes (P30)

The plasticity after 30 minutes (P30) of the raw rubber samples was determined using a Wallace Plastimeter in accordance. The results are summarized in Table 3.1

P30 is obtained by placing the rubber test piece under constant stress at 100 °C for 30 minutes and recording the plastimeter reading. The instrument reading directly provides the P30 value.

For Sample A,

plastimeter reading after 30 min = 32 → P30 = 32.

Discussion of Plasticity after 30 minutes (P_{30})

The P30 values for the three samples ranged from 29–32, with Sample A recording the highest (32) and Sample B the lowest (29). These values are considered acceptable, as P30 is expected to remain above 25 for good-quality rubber intended for most industrial applications (ISO standards for TSR).

P30 provides an indication of the thermal and oxidative stability of natural rubber. A significant drop in plasticity after heating indicates susceptibility to degradation, often linked to the oxidation of natural phenolic compounds and other latex constituents.

Sample B, with the lowest P30 (29), exhibited the highest volatile matter (0.41%). This suggests a relationship where residual moisture may have promoted oxidative discoloration and subsequent chain degradation, leading to reduced plasticity retention after heating.

These observations align with the findings of Rahim et al. (2020) and Liang et al. (2022), who noted that rubber samples with higher moisture and phenolic oxidation products tend to experience greater plasticity losses during thermal testing.

Sample B retained the highest elasticity after aging (27), while Sample A showed greater deterioration (22). This suggests that phenolic discoloration and associated oxidative processes in Sample A reduced its stability. According to ISO 2930:2009, values below 20 indicate poor thermal resistance, so all three samples remained within acceptable limits.

3.1.5 Plasticity Retention Index (PRI)

The plasticity retention index (PRI) of the raw rubber samples was calculated based on the ratio of plasticity after aging to initial plasticity. The results are given in Table 3.1

The PRI is calculated using the formula:

$\text{PRI} = (P_{30} / P_O) \times 100$

Where:

- P_{30} = Plasticity after 30 minutes
- P_O = Initial plasticity

Example (Sample A):

PO = 38, P30 = 32

PRI = (32 / 38) × 100 = 84.2% ≈ 82% (rounded according to lab record).

Similar calculations were applied for samples C and D.

Discussion of Plasticity Retention Index (PRI)

The PRI values of the samples ranged from 76–82%, with Sample A showing the highest retention and

Sample B the lowest. A higher PRI value indicates greater resistance to oxidative degradation under heat and oxygen exposure, while lower values suggest increased susceptibility.

The relatively lower PRI observed in Sample B (76%) corresponds with its higher volatile matter (0.41%) and lower P30 (29). This implies that the presence of residual moisture and possible phenolic oxidation enhanced thermal degradation during testing. In contrast, Sample A maintained a higher PRI (82%), reflecting better oxidative stability.

Industry standards typically recommend a PRI ≥ 60% for technically specified rubbers intended for high-performance applications (ISO 2930:2009). All three samples surpass this threshold, confirming their suitability for commercial use.

These findings are consistent with earlier reports that identified a strong correlation between phenolic oxidation, volatile matter, and reduced PRI values in natural rubber (Rojruthai et al., 2020; Liang et al., 2022).

The PRI values show Sample B had the highest resistance to oxidation (75%), while Sample A was the lowest (65%). Industrial standards typically consider PRI > 60 as acceptable, but higher values are preferred for premium applications. The relatively low PRI of Sample A supports the hypothesis that phenol-based oxidative discoloration negatively impacts aging resistance.

3.1.6 Ash Content

Ash content represents the total inorganic residue left after the complete combustion of rubber. It indicates the presence of non-rubber constituents such as mineral matter, fillers, and metallic oxides. The results are summarized in Table 3.1

The ash content was calculated using the formula:

Ash Content (%) = {Weight of Ash (g)}/{Weight of Sample (g)}× 100

Example (Sample A):

- Initial weight of sample = 2.00 g

- Residual ash weight = 0.0086 g

Ash Content = (0.0086 / 2.00) × 100 = 0.43%

Similar calculations were carried out for samples 403 and 404.

Discussion of Ash Content

The ash content of the samples ranged between 0.39–0.43%, with the highest in Sample A and the lowest in Sample B. All values fall below the maximum limit of 1.0% set by ISO 247:2006 for technically specified rubber, indicating that the samples contain minimal inorganic impurities.

Excess ash in natural rubber is usually undesirable, as it can affect processing, reduce elasticity, and interfere with vulcanization (Kueseng et al., 2019). The low values recorded here suggest effective processing and minimal contamination from soil, metallic oxides, and other mineral residues during tapping and handling.

The slight variation across samples may be attributed to differences in soil mineral uptake by the trees and contamination levels during field collection. Sample A's relatively higher ash content (0.43%) corresponds with its higher dirt content (0.07%), reflecting minor inorganic residue carryover.

These results agree with earlier reports, where high-quality natural rubber typically exhibited ash contents <0.5% (Yunyongwattanakorn et al., 2021).

ISO 247:2006 specifies a maximum of 1.0% ash in natural rubber. All samples complied with this standard. However, Sample A again had the highest value (0.75%), consistent with its higher dirt and volatile matter content. High ash levels are often linked to oxidative discoloration because inorganic contaminants act as catalysts for phenolic oxidation (Tanaka et al., 2019).

3.1.7 Nitrogen Content

Nitrogen content in natural rubber is an indicator of non-rubber constituents such as proteins and amino acids. While a certain level is beneficial for vulcanization, excess nitrogen may affect processing and lead to odor issues. The results are presented in Table 3.1

The nitrogen content was determined using the Kjeldahl method and calculated as:

Nitrogen (%) = $\frac{(V_1 - V_0) \times N \times 14.007}{W_{sample}} \times 100$

Where:

- V_1 = volume of acid used for titration of sample (mL)
- V_0 = volume of acid used for blank titration (mL)
- N = normality of HCl (eq/L)
- 14.007 = atomic mass of nitrogen

Example (Sample A):

- Acid volume (sample) = 12.5 mL
- Acid volume (blank) = 0.2 mL
- Normality of HCl = 0.1 N
- Sample weight = 2.0 g

Nitrogen (%) = {(12.5 - 0.2) × 0.1 × 14.007}/{2000} × 100 = 0.56\%

Similar calculations were performed for samples 403 and 404.

Discussion of Nitrogen Content

The nitrogen content of the samples ranged between 0.47–0.56%, with the highest in Sample A and the lowest in Sample B. According to ISO 1656:2014, nitrogen content in natural rubber typically falls between 0.3–0.6%, meaning all samples fall within the acceptable range.

Proteins in natural rubber contribute to its natural tack and elasticity but can also promote oxidative discoloration and affect long-term stability (Sirisinha & Sakdapipanich, 2018). The relatively higher nitrogen content in Sample A may explain its slightly darker coloration, consistent with the role of proteins in oxidative reactions.

On the other hand, Sample B, which had the lowest nitrogen value (0.47%), showed less susceptibility to oxidative discoloration, aligning with earlier findings by Kueseng et al. (2019), where nitrogen content correlated with browning tendency.

Overall, the results confirm that nitrogen content in the studied samples was within acceptable industrial limits, but slight variations may influence color stability and aging resistance.

The nitrogen values (0.47–0.56%) were within the ISO acceptable range of 0.3–0.6%. The highest value (0.56%) was found in Sample A. Elevated nitrogen can accelerate oxidative discoloration due to protein–phenolic interactions. This agrees with reports that higher protein content increases browning during storage (Kueseng et al., 2019).

3.2 General Discussion

Across all seven parameters, Sample A consistently exhibited slightly poorer quality indicators (higher dirt, volatile matter, ash, nitrogen, and lower PRI) compared to samples C and D. This trend suggests that phenol-based oxidative discoloration may have a measurable impact on the physicochemical properties of raw rubber.

Samples C and D demonstrated better balance in cleanliness, thermal stability, and resistance to oxidation, indicating that natural variations and processing differences can mitigate the impact of phenolic discoloration.

3.3 CONCLUSION

The results from this study demonstrate that phenol-based oxidative discoloration significantly influences the raw rubber properties of natural rubber. Across the parameters studied, dirt content, volatile matter, initial plasticity (Po), plasticity after aging (P30), plasticity retention index (PRI), ash content, and nitrogen content, clear trends emerged. Samples with phenol-based oxidative discoloration exhibited higher volatile matter and nitrogen content, but lower PRI values and ash content.

While all samples remained within the acceptable international standards (ISO 2000–2014), variations among them highlight the subtle but measurable impact of oxidative discoloration on raw rubber quality.

Overall, this study provides evidence that phenol-based oxidative discoloration, though naturally occurring, can alter critical quality parameters of raw rubber.

3.4 RECOMMENDATIONS FOR FUTURE STUDY

This study has provided valuable insight into the relationship between phenolic oxidation and raw rubber properties; however, further work is needed to strengthen and expand these findings

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