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Synthesis and utilization of rice husk cellulose nanocrystals as biocomposite film reinforcement for edible packaging application

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Abstract
The high-value application of rice husk, a renewable, easily accessible, and plentiful agricultural waste, demands further study. The goal of this research is to create rice husk cellulose nanocrystals (CNC) reinforced carboxyl methylcellulose (CMC)-gelatin film using solution casting method for use in edible packaging. CMC-gelatin nanocomposites reinforced with CNC through solution casting technique were successfully fabricated. Investigations were conducted into how CNC content affected the morphological, mechanical, water vapor permeability (WVP), degradability, and thermal characteristics of carboxyl methylcellulose-gelatin-CNC films. The results of the scanning electron microscopy showed a needle-like form with dimensions of 81 to 286 nm in length, 8 to 21 nm in diameter, aspect ratio of 17, and crystallinity index of 0.82. The WVP of the films decreased from 3.10 x 10^{-6} g/m.h.Pa to 2.32 x 10^{-6} g/m.h.Pa. The tensile strength of the nanocomposite films increased from 6.83 MPa to 7.80 MPa, elongation at break reduced from 81.50% to 63.75% while thickness increased from 0.13 um to 0.25 um by CNC incorporation. The degradability of the nanocomposite films also reduced from 0.24 wt% to 0.21 wt%, while the thermal stability of the nanocomposite films increased from 283^{o}C to 363^{o}C. The result indicates clearly the vantages in the use of CNC reinforced CMC-gelatin to replace the extensive use of petroleum based material.

Keywords: Rice husk, cellulose nanocrystal, Nanocomposite, Packaging

1. Introduction

Despite the fact that petroleum-based synthetic plastics are frequently used for packaging foods, the rising environmental pollution and issues with food safety brought on by these materials have forced researchers to explore for secure biodegradable alternatives (Youssef and El-Sayed, 2018). Since edible and biodegradable films have desirable advantages to prevent moisture loss, aroma loss, solute transport, water absorption in the food matrix, and oxygen penetration, they could be appealing replacements for non-biodegradable plastic packaging materials in the food industry (Salgado *et al.,* 2015; Cazón *et al.,* 2017). There have recently been significant attempts made to produce edible and biodegradable films using natural bipolymers (Nouraddini *et al.,* 2018; Azeredo *et al.,* 2016). Natural polymers such as polysaccharides (Nešić *et al.,* 2019), proteins (Maryam Adilah *et al.,* 2018), and lipids (Mohamed *et al.,* 2020) are considered as potential resources for preparing biodegradable packaging films due to their biodegradability, non-toxicity, easy accessibility, and renewable properties. Polysaccharides may be the most promising biopolymer options because of their alluring mix of thermoplastic behavior, quantity, and affordability (Cazón *et al.,* 2017).

The anionic polysaccharide CMC, which has been widely utilized as a thickening or stabilizer in the food processing sectors (Akhtar *et al.,* 2018), is created by adding CH_2COOH groups to the cellulose molecular chain. CMC has generated a lot of interest as an edible and biodegradable film material for food packaging because of its superior film-forming ability, harmless and low cost (Roy and Rhim, 2020; Zabihollahi *et al.,*2020). The CMC film demonstrated good transparency and efficient oxygen and carbon dioxide barrier characteristics, according to numerous tests (Zabihollahi *et al.,* 2020). However, the mechanical and water vapor barrier qualities of CMC film are very subpar, which limits its potential use in the food packaging industry (de Melo Fiori *et al.,* 2019). Although adding hydrophobic lipids to the CMC matrix can lessen the permeability to water vapor, this will have a negative impact on the mechanical and optical characteristics of the films (de Melo Fiori *et al.,* 2019). To create a CMC-based composite film, one of the most popular ways to achieve this is to combine nanoparticles such as nanoclay (He *et al.,* 2020), nanocellulose (Zabihollahi*et al.,* 2020; Oun and Rhim, 2015), chitin nanocrystals (Oun and Rhim, 2017), and nanometal oxides (Roy and Rhim, 2020; Achachlouei and Zahedi, 2018) with the CMC matrix. Nanocellulose can create potent hydrogen bonds with biopolymers in the polymer matrix, which will enhance the mechanical properties of the composite film. Additionally, they can act as physical barriers for water molecules, forcing them to take a circuitous route through the polymer matrix and slowing the flow of water through composite films (Sun *et al.,* 2018). Traditionally, renewable cellulosic feedstock is hydrolyzed with sulfuric acid to produce nanocellulose (Xie *et al.,* 2018). Agricultural waste is a desirable source of cellulosic feedstock because it doesn't endanger food supplies and boosts the rural economy. Agricultural waste, including rice husk, pineapple peel, banana peel, peach palm extraction waste, sugarcane bagasse, wheat straw, and soy hull, has recently been investigated as a potential biomass source for making nanocellulose. Rice is a significant agricultural crop for humans due to their high protein content (23–33%) (Lan *et al.,* 2018). Human consumption or food processing facilities produce a significant amount of trash made up of rice husk, most of which is abandoned and burned and inevitably leads to secondary environmental damage.

A high-value utilization strategy for this waste biomass is provided by the discovery that rice husks contain silica (15–28 weight percent) and lignocellulose (75–90 weight percent), which is further divided into cellulose (35–40 weight percent), hemicellulose (15–20 weight percent), and lignin (20–25 weight percent) (PREGA, 2014). Gelatin is one of the best biopolymers ever used in the production of films. A biopolymer made from collagen is gelatin. Since gelatin has a hydrophilic character, mechanical properties of gelatin gels are very sensitive to temperature variations, the previous thermal history of the gels, and the amount of time elapsing. These gels exist over only a small temperature range, the upper limit being the melting point of the gel, which depends on gelatin grade and concentration (Gómez-Guillén *et al.,* 2019). Gelatin is used to coat capsules in the pharmaceutical business, and data from the food industry indicate that gelatin has antioxidant properties when used to cover meat during storage (Neenu *et al.,* 2022).

At low relative humidity, gelatin can create films with advantageous optical, mechanical, and protective qualities against gas, oxygen, and odor. Cellulose nanocrystals (CNCs) are crystalline particles produced as a result of acid hydrolysis treatment, which removes the amorphous region leaving the crystalline region (Neenu *et al.,* 2022). According to Bagde and Nadanathangam, (2019), CNC has special qualities like cheaper price, nontoxicity, higher thermal stability, optical clarity, and biodegradability. However, due to its exceptional thermal and mechanical properties, CNC is frequently used as a reinforcing material in polymer composites (Li *et al.,* 2022).

Different biodegradable polymers that would otherwise be inappropriate for many applications have their physicochemical, thermal, and insulating properties significantly improved by CNC (Kian *et al.,* 2019). Additionally, CNC's high elasticity modulus (roughly 150 GPa) (Amara *et al.,* 2021), ultra-light weight ($1.6 g/cm^3$) (Daicho *et al.,* 2020), biodegradability (Kargarzadeh *et al.,* 2018), and biocompatibility (Huang *et al.,* 2018) are the main factors that promote its use as a reinforcement agent in composite manufacturing. According to Raza and Abu-Jdayil, (2022) and Xiong *et al.,* (2021), CNCs are typically produced from agricultural waste biomass such as rice husk, cotton stalks, corncobs, wheat straw, coconut husks, maize straws, and pea hull fibers. Although rice husk is a cheap and plentiful biomass resource, there have only been a few numbers of studies on the synthesis of nanocellulose from rice husk waste that have been published in the literature. In order to better utilize and dispose of this agricultural waste, the current study attempted to isolate nanocellulose from waste rice husk and evaluate its reinforcing capabilities for CMC/gelatin film. To accomplish these goals, cellulose nanocrystals (CNC) were initially extracted from rice husk utilizing the alkaline treatment method with sodium hydroxide (NaOH), bleaching with acetic acid and sodium chlorite, and sulfuric acid hydrolysis procedure. The generated CNC was then added to the CMC matrix as a reinforced additive to create composite films made of CMC, gelatin, and CNC using the

solution casting method. The impacts of CNC content on the morphological, mechanical, water vapor permeability, degradability, and thermal properties of CMC/gelatin/CNC based composite films were examined.

2.0 Materials and Methods

2.1 Materials

The ingredients were obtained from Onitsha Bridge-head Chemical Market, Anambra State, Nigeria. The analytical grades of carboxylmethyl cellulose (molecular weight, 115 000 g/mol; degree of polymerization, 1700-1800), gelatin (bloom strength 185), glycerol (98%), sulphuric acid (99%), sodium hydroxide, sodium chlorite, acetic acid (98%), benzene (99%), ethanol (99%), calcium chloride (96%) and potassium sulphate (99%), distilled water, rice husk from Nwaokeanyanwu Agricultural Farm in Awka.

2.2 Extraction of rice husk cellulose

Miri *et al.,* (2015) modified a similar method for extracting cellulose to obtain the desired results. The fibers were dried, crushed, and sieved after being rinsed with distilled water for one hour at 60°C with mechanical stirring.

2.2.1 Alkaline pretreatment of the fibre: The prewashed fibers were immersed in a 4 weight percent sodium hydroxide solution at a fibre to liquor ratio of 1:20 for two hours at 80°C while being mechanically stirred. The fiber were then thoroughly rinsed with distilled water and dried in an oven at 100°C to a constant weight.

2.2.2 Bleaching pretreatment of the fibre: Following drying, 1g of fibre was added to a solution containing 65ml of water, 0.5ml of acetic acid, and 0.6g of sodium chlorite, which was then heated to 80°C for 4 hours until the fibre becomes white. After this process, what is left is called cellulose, which is stored in desicator till further analysis.

2.3 Preparation of cellulose nanocrystals (CNC)

CNC was obtained by the controlled sulphuric acid hydrolysis. Dried cellulose was mixed with 64 wt% sulphuric acid (H_2SO_4) solution at 1g cellulose per 8.75 ml acid with strong mechanical stirring at 45ºC for 30 minutes (Csiszar and Nagy, 2017). To dilute and stop the reaction, 10-fold solution of deionized water was utilized. The supernatant was removed after centrifuging the chilled suspension. The solid content was Once again diluted, thereafter centrifuged until the supernatant turned turbid. The turbid supernatant was dialysed for a week using deionized water, resulting in a pH of roughly 6.5-7.

2.4 Film preparation

The film was made using the same solution casting technique as Mohammadi *et al.,* (2019). With steady stirring, distilled water was heated to 90°C and used to dissolve a predetermined weight of carboxylmethyl cellulose for 30 minutes. Gelatin with a given weight was dissolved in distilled water for 30 minutes at 80°C. Gelatin and carboxylmethyl cellulose were combined and constantly swirled for 30 minutes at 90°C. At room temperature, the necessary amount of cellulose nanocrystal was added to glycerol, which was then stirred and heated to 90°C before being added in drops to the gelatin-carboxylmethyl cellulose solution. The resulting solution was then placed in a petri plate and air dried for 48 hours to allow the solvent to slowly evaporate. The films were kept in a desiccator until examination after the water had entirely evaporated.

2.5 Experimental Design using One-Factor-At-A-Time (OFAT) Design

One-Factor-At-A-Time (OFAT) Design was used to study the effect of concentration of carboxylmethyl cellulose, gelatin and cellulose nanocrystals on the dependent factors (tensile strength, elongation at break and thickness). Equal concentrations of gelatin and carboxylmethyl cellulose was chosen to determine the overall effects of three different CNC concentration (0%, 5% and 10%) on dependent variable (tensile strength, elongation at break and thickness).

2.6 Mechanical test

The M500-25T universal mechanical testing machine was used to conduct the tensile test in accordance with ASTM D882. The films were divided into strips of 100 mm by 20 mm. Before testing, the thickness of each specimen was measured. A digital caliper (Traceable® Digital Calipers 6in, Fischer Scientific) was used to measure the thickness of the film. Test conditions were a 60mm grip gap, a 5N pretension, and a 50mm/min cross head speed. Prior to testing, the samples were stored for 24 hours in a desiccator with calcium chloride desiccant.

$$\text{Tensile Strength} = \frac{Load(N)}{Thickness(mm) \times Width(mm)}$$

$$\text{Elongation at break}(\%) = \frac{Displacement\ at\ break}{Guage\ length} \times 100$$

2.7 Scanning electron microscopy (SEM)

Utilizing the VEGA 3 TESCAN, scanning electron microscopy was carried out. It was employed to observe the biocomposites' cracked surface. The films were cryofractured in liquid nitrogen, placed on silica gel, and kept at 25°C over a bronze stub coated in a gold-palladium alloy. Samples were observed using an accelerating voltage of 20Kv.

2.8 Water vapour permeability (WVP)

With a few adjustments, the standard method ASTM E96-E95 was used to determine the water vapour permeability of the biocomposite films. To carry out the test, glass bottles with known diameter and depth were utilized. In order to keep the relative humidity (RH) within a glass bottle at zero percent, four grams of anhydrous calcium chloride ($CaCl_2$) were added. Glass bottles were placed in a desiccator containing potassium sulphate (K_2SO_4) with a RH of 98% and a temperature of 25°C, coated with a complex biocomposite film, and weighted. Every 24 hours, the process of weighting was conducted. As a function of time, the glass bottle's weight variation was being recorded. Linear regression was used to calculate slopes (weight change versus time). The slope of the straight line (g/h) divided by the area of the glass bottle mouth (m^2) yielded the water vapour transfer rate (WVTR). Finally, as published by (EL Miri *et al.,* 2015), the water vapour permeability (gm/m^2hPa) was determined using the following equation.

$$\text{WVP} = \frac{WVTR}{S(R_1 - R_2)}X$$

Where X is the film's thickness in meters, S is the saturated water vapour pressure (Pa) at 25 degrees Celsius, R_1 and R_2 are the relative humidity in the climate chamber and the glass bottle, respectively.

2.9 Soil degradation

With a few minor modifications, the soil degradation tests were carried out in accordance with Salisu *et al.,* 2014. The test films were weighted and buried at a depth of 6 cm in a sand-filled bin. At intervals of five days, the samples were carefully removed from the dirt, and the film was cleaned and pat dried. The deterioration rate of the film was calculated by estimating the weight loss of the film over time after it had completely dried.

2.10 Thermogravimetric analysis (TGA)

A TGA Q50 thermogravimetric analyzer was used to perform the thermogravimetric analysis. To learn more about how cellulose nanocrystals degrade, thermogravimetric analysis was used. The samples were heated under nitrogen from room temperature to 500^{o}C at a rate of 10^{o}C/min.

3.0 Results and Discussions

Table 3.1: **Mechanical properties of nanocomposite films**

	Factor 1	Factor 2	Factor 3	Response 1	Response 2	Response 3
Run	**A:GELATIN (g/100g)**	**B:CMC (g/100g)**	**C:RH-CNC (wt %)**	**T.S (MPa)**	**E.B (%)**	**THICKNESS (UM)**
1	7.00	3.00	0.00	6.83	81.50	0.13
2	7.00	3.00	5.00	7.80	72.81	0.17
3	7.00	3.00	10.00	6.57	63.75	0.25

RH= Rice husk, T.S= Tensile strength, E.B= Elongation at break, UM= Micrometer

In addition to assessing a film's capacity to keep its integrity during packing applications, a film's mechanical qualities also reflect its capacity to endure mechanical damage during handling, storage, and shipment. One of the key metrics for determining how much stress a nanocomposite film can sustain before breaking is its tensile strength. Comparing the CNC addition to the 0% CMC/gelatin nanocomposite film, the tensile strength was dramatically increased. The tensile strength of the 0% CMC/gelatin nanocomposite film was 6.83 MPa, and the addition of 5 wt% CNC content raised it to 7.80 MPa. Tensile strength dropped to 6.57 MPa after 10 wt% CNC material was added as shown in Table 3.1.The formation of intermolecular hydrogen bonds between CNC molecules and matrix molecules was primarily responsible for the increase in tensile strength of CMC/gelatin/CNC nanocomposite film, and this beneficial interaction between CMC/gelatin and CNC was reduced at high CNC content (10 wt%) due to CNC aggregation formation (Savadekar and Mhaske, 2014). Additionally, as the percentage of CNC increased, the flexibility of the composite films as measured by elongation at break dramatically decreased. The 0 wt% CNC reinforced CMC/gelatin nanocomposite film's elongation at break was 81.50%. After adding 5 wt% and 10 wt% of CNC to the CMC/gelatin matrix, respectively, the elongation at break dropped to 72.81% and 63.75%. According to Chen *et al.,* (2019), the enhanced stiffness of the composite film restricts the motion of the CMC/gelatin matrix molecular chains, hence limiting the elongation at break. The thickness of the 0 wt% CNC reinforced CMC/gelatin nanocomposite film was 0.13 um; with the addition of 5 wt% and 10 wt% CNC, it increased to 0.17 um and 0.25 um respectively. Due primarily to the increased solid content of the nanocomposite films, the thickness of the nanocomposite films rose as the content of CNC increased. This effect is in line with a study by Reddy and Rhim, (2014), who discovered that as cellulose nanocrystal content increased, so did the thickness of the agar/cellulose nanocrystal film.

3.2 **Morphological analysis**

In order to investigate the dispersion of CNC in the CMC/gelatin matrix and the compatibility between CMC/gelatin and CNC, the surface morphology of the various CMC/gelatin/CNC nanocomposite films was examined by scanning electron microscopy. The surface of the 0 wt% CNC reinforced CMC/gelatin was uniform, smooth, and free of air bubbles as illustrated in Fig. 3.1. The CMC/gelatin nanocomposite films continued to have uniformly smooth surfaces up until 5 wt% CNC was added, at which point deep cracks and air bubbles appeared, as illustrated in Fig. 3.2. These findings suggested that the CNC had good compatibility with CMC/gelatin and was evenly dispersed across the continuous CMC/gelatin matrix (Oun and Rhim, 2017). However, after adding 10 wt% CNC as shown in Fig. 3.3, some agglomeration in the CMC/gelatin/CNC nanocomposite film developed. This was likely caused by the agglomeration or inhomogeneous dispersion of CNC in the CMC/gelatin matrix at higher CNC concentrations (Oun and Rhim, 2015). A similar finding was also made with a composite film made from gellan gum and nanocellulose that was extracted from pineapple peel (Dai *et al.,* 2018).

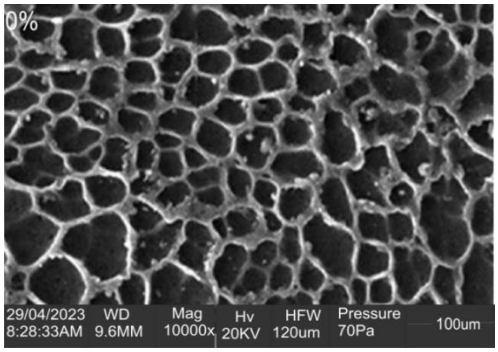


Fig. 3.1: 0 wt% CNC

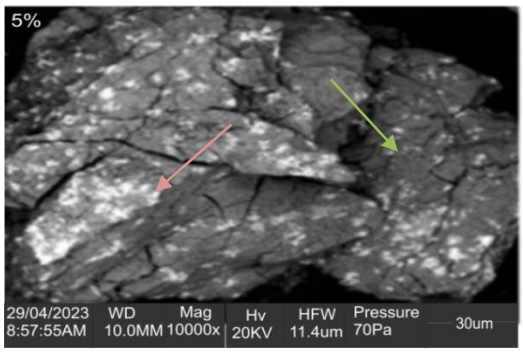


Fig.3.2: 5 wt% CNC

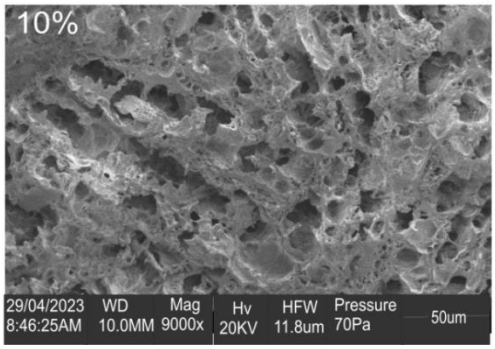


Fig. 3.3: 10% CNC (Green arrow=deep crack, red=air bubble)

3.3 **Water vapor permeability (WVP)**

For food packaging applications, the water vapor permeability quality of the film is extremely important, and it should be as low as feasible (Saha *et al.*, 2016). Fig. 3.4 depicts the water vapor permeability of the CNC reinforced CMC/gelatin nanocomposite films. The water vapor permeability of 0 wt% CNC reinforced CMC/gelatin nanocomposite film was 3.10 x 10^{-6}g/m.h.Pa, and it dramatically dropped when CNC was added to the CMC/gelatin matrix. In comparison with 0 wt% CNC reinforced nanocomposite film, the water vapor permeability of 5 wt% CNC reinforced CMC/gelatin nanocomposite film dropped to 2.69 x 10^{-6}g/m.h.Pa. The fact that the CNC was evenly distributed throughout the CMC/gelatin matrix may have contributed to the water vapor permeability's decline by creating a convoluted pathway for water molecules to diffuse and lengthening their trip through the films (Kanmani and Rhim, 2014). However, the addition of 10 wt% CNC reinforced CMC/gelatin nanocomposite film resulted in a minor drop in the water vapor permeability of the nanocomposite film to 2.32 x 10^{-6}g/m.h.Pa, which was caused by the aggregation of CNC particle in the polymer matrix.

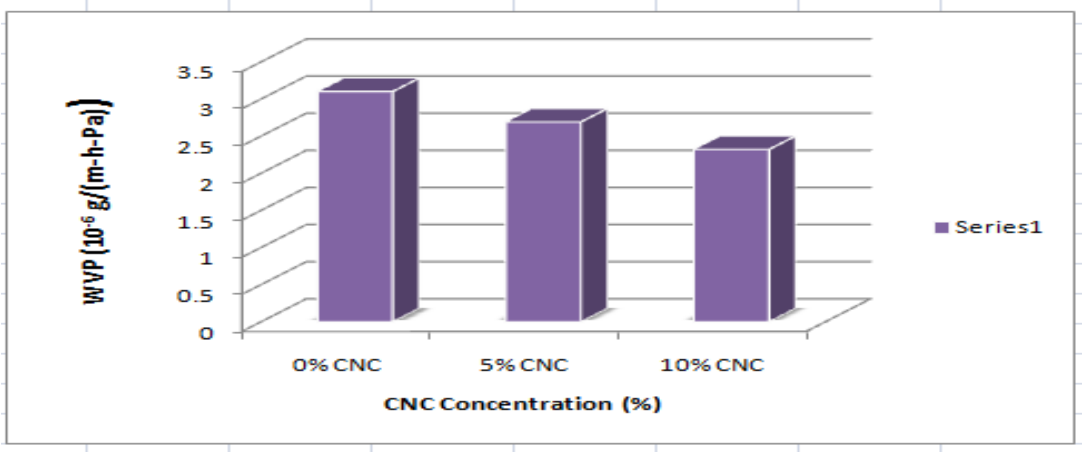


Fig. 3.4: Water vapour permeability of nanocomposite films

3.4 **Soil burial test (degradability)**

The 10 wt% CNC reinforced CMC/gelatin nanocomposite film sample decomposed more quickly than other samples, even though complete biodegradation was not observed during the study time. The results are shown in Figure 3.5. Within the first 25 days, there was a noticeable rapid rate of degradation, with the 10 wt% CNC reinforced film surprisingly degrading faster than the 5wt% CNC reinforced film. But after 30 days of soil burial test, the biodegradation of the nanocomposite was nearly the same for the unreinforced (0 wt% CNC) and reinforced (5 wt% and 10 wt% CNC) CMC-gelatin films. Then after 50 days of soil burial test, unreinforced films had a degradability of 0.24%, while 5 wt% CNC and 10 wt% CNC reinforced films saw weight losses due to biodegradation of 0.23% and 0.21% respectively. Without a doubt, 10 wt% CNC reinforced film may have distributed in the CMC-Gelatin matrix better than 5 wt% CNC reinforced film, creating a better 3-dimensional network structure. In general, addition of CNC decreases the rate of degradation of the nanocomposite film from 0.24% down to 0.21%. Even though the rate of degradation for unreinforced films had significantly decreased over the previous four days, they all exhibited the potential for further degradation if the degradation time was extended. It's been verified already (Danjaji *et al.*, 2015).

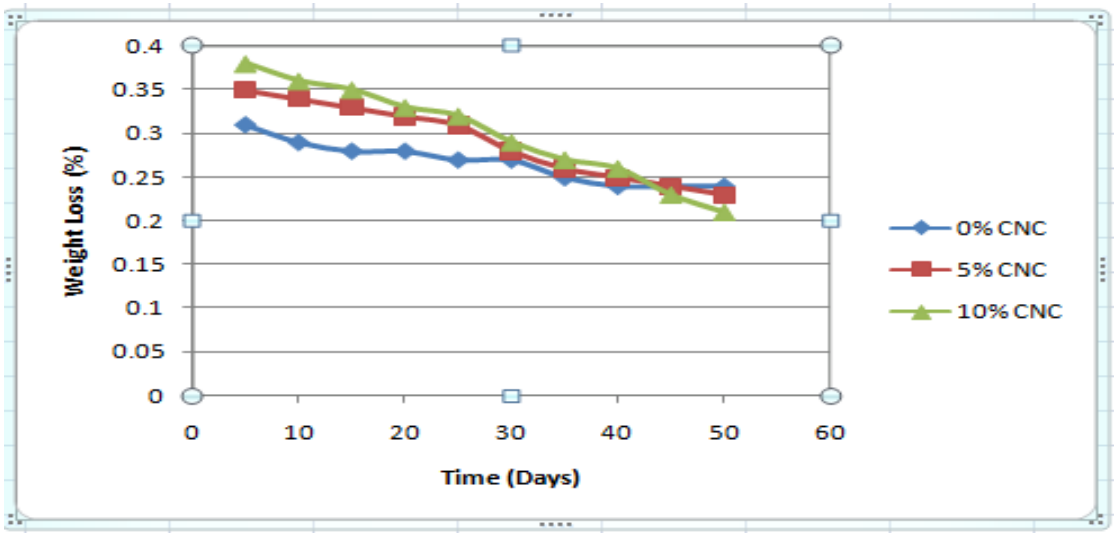


Fig. 3.5: Soil degradation of the nanocomposite films

3.5 **Thermogravimetric analysis (TGA**)

Characterizing a material's processing temperature range and uses requires analysis of its thermal properties. Thermogravimetric analysis (TGA) and differential thermal analysis (DTA) were used to analyze the thermal behaviors of the various CMC/gelatin/CNC composite films, and the findings are displayed in Fig. 3.6. Two primary stages of decomposition were visible in the thermal degradation curve. In the first stage of decomposition, 0 wt% CNC started to break down at 283 ºC and ended up at 406°C, 5% of the CNC started to break down at 307°C and ended up at 363°C, and 10% of the CNC started to break down at 288°C and ended up at 355°C.This can be the result of the polymer losing both bound and free water. All of the breakdown peaks of the nanocomposite migrated to a little higher temperature with the addition of CNC, according to the DTA, which represents the second stage of weight loss. By serving as an insulator and mass transport barrier to the volatile products produced during decomposition CNCs slightly improved the thermal stability of the nanocomposite. The degradation temperature of the 0 wt% CNC nanocomposite was 325 ºC, and it increased to 330ºC and 340ºC upon addition of 5wt% and 10 wt% CNC, respectively (Yadav *et al.*, 2019). The breakdown of the polymer backbone may be responsible for the weight loss in both polymers during the second stage.

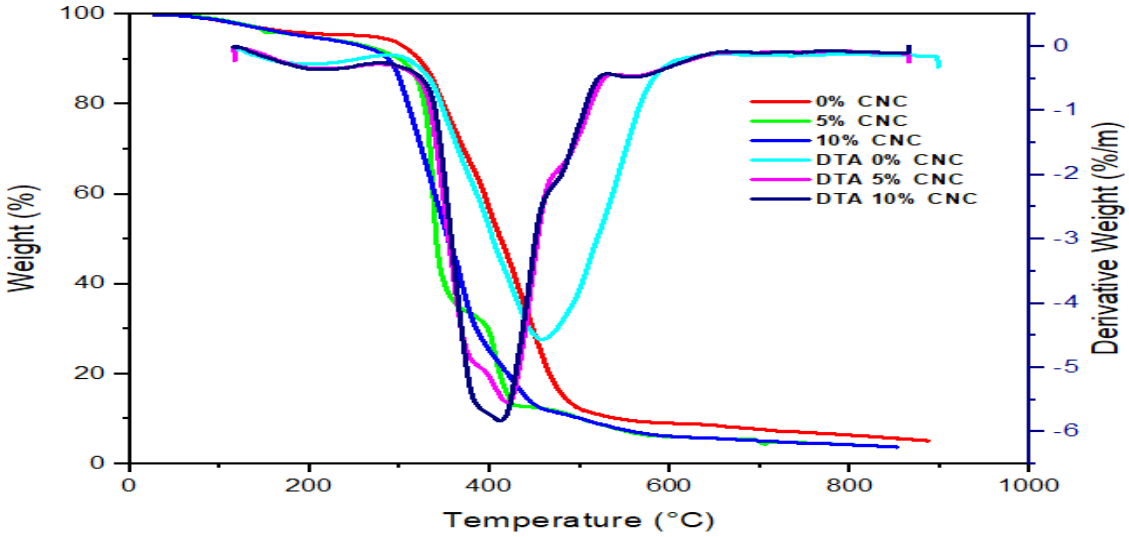


Fig. 3.6: Thermal decomposition of 0wt%, 5 wt% and 10 wt% CNC

4.0. Conclusion

One of the most well-liked crops and a valuable commodity all over the world is rice. However, once the rice seed harvest is complete, rice husk often degrades into biomass waste. As a result, scientists have begun exploiting rice crop fiber to create a variety of products with added value. Using an acid hydrolysis technique, bleaching pretreatment, and alkaline pretreatment, cellulose nanocrystals from rice husk were extracted. The solution casting approach was used to successfully prepare the CNC/Gelatin/CMC nanocomposite films. The research investigated how varied CNC loadings (0 wt%, 5 wt%, and 10 wt%) affected the reinforcing of CMC/Gelatin nanocomposite film. The CMC/gelatin/CNC composite film's morphological, mechanical, water vapor permeability, degradability, and thermal properties were determined. Scanning electron micrographs showed that the CNCs were distributed evenly and adhered to the CMC/Gelatin matrix well. The mechanical properties of the nanocomposite films were also found to be improved by the addition of CNC to the CMC/Gelatin matrix, with 5 wt% CNC exhibiting the maximum tensile strength and thickness and 0 wt% CNC exhibiting the highest elongation at break. Additionally, the water vapour permeability and degradability of the nanocomposite films are reduced when cellulose nanocrystals are added. The thermal stability of the CMC/Gelatin film reinforced with 5 wt% CNC was higher than that of the CMC/Gelatin film reinforced with 0 wt% CNC. In comparison to CMC/Gelatin film with 0 wt% reinforced CNC, all reported analyses indicate that the addition of CNC to the CMC/Gelatin film had the best attributes. Therefore, the future lies in increasing the water vapour permeability, degradability, and thermal properties of the nanocomposite films through additional study and technical improvements.

5.0 Recommendation

Based on the findings, the following recommendations were made,

- The use of CNC reinforced packaging film from rice husk fibres should be encouraged to reduced agricultural waste pollution and create waste to wealth.
- The use of rice husk fibres for reinforcement by industries should be encouraged.

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