IJBPAS, February, 2025, 14(2): 813-823

ISSN: 2277–4998

International Journal of Biology, Pharmacy and Allied Sciences (IJBPAS)

'A Bridge Between Laboratory and Reader'

www.ijbpas.com

TOWARDS A GREENER FUTURE: INNOVATIVE SYNTHESIS OF BIODEGRADABLE POLYMER FROM *Citrullus Lanatus* PEELS AND WASTE NEWSPAPER SHEETS

THAKER TIRTH*, PANCHANI DIPEN, RAULJI MAYURDHVAJSINH, GADHAVI ASHOKDAN

Department of Chemistry, Parul Institute of Applied Sciences, Parul University, Vadodara-391760, Gujarat, India

***Corresponding Author: Dr. Tirth H. Thaker: E Mail: tirth6582@gmail.com**

Received 4th Nov. 2023; Revised 5th Dec. 2023; Accepted 2nd May 2024; Available online 1st Feb. 2025

https://doi.org/10.31032/IJBPAS/2025/14.2.8696

ABSTRACT

Petrochemical plastic has overrun the planet in recent years. Petroleum-based plastics are very common in our lives. The growing concern over environmental pollution caused by conventional plastics has led to an urgent need to develop sustainable and biodegradable alternatives. Citrullus lanatus (watermelon) peels can be utilized in bioplastic production with some additives like starches, glycerine, waste newspaper pulp, citric acid, etc. This newly created bioplastic may be used as a biodegradable food packaging material to improve food quality and safety and for normal packaging. Biodegradable plastic properties are studied by FTIR, XRD and different types of biodegradation studies and this biodegradable plastic also shows potent antibacterial activity.

Keywords: Biodegradable, *Citrullus lanatus* peels, starch, glycerine

INTRODUCTION

Plastic consumption in the world is more than 200 million metric tons per year, of which half is global plastic usage. Most of them are used only once and are non-biodegradable and non-recyclable food packaging waste, harming the water and landfills. So, there is an urgent need for biodegradable polymer film, which has no negative effect on the environment and is also readily available and affordable for everyone **[1]**. The processing of fruits and vegetables generates enormous amounts of garbage each year which pollutes the environment so, it can be reused in innovative ways. *Citrullus lanatus* is a member of the Cucurbitaceae family. Due to their high pectin, starch and cellulose watermelon pills have been utilized to extract natural polymers **[2, 3]**. Its peel is mostly used in animal feed supplements and fertilizer and 90 percent of it is released into the environment **[4]**. Citrullus lanatus by-products have been shown in several studies to have bio-medical qualities, which makes them an excellent natural source for pharmaceuticals **[5]**.

Due to the inclusion of bioactive substances like L-citrulline and phenolic substances, including benzoic acid, vanillin, phenol, and alkaloids, which are important active substances, it has a strong antioxidant **[6]**, antimicrobial **[7]**, cosmetic, gastronomic, and medicinal applications **[8]**. The biodegradable polymer of watermelon pills and newspaper pulp makes it possible to reduce organic wastage. It can be used to make sheets and it can be moulded into food packaging material for preservation and safety **[9]**.

Accordingly, the primary goals of this research endeavor revolved around the formulation of biodegradable films sourced from a composite of *Citrullus lanatus* peel combined with newspaper pulp. Examination of biodegradable films were carried out using different characterization techniques.

EXPERIMENTAL

Material

Each Chemical was purchased from Sigma-Aldrich. On a Fourier transform Digilab Scimitar Series spectrometer, IR spectra were recorded. X-ray diffractometer (XRD) of the film was recorded in advance D8, Bruker spectrometer.

Preparation of *Citrullus lanatus* peel powder (MR-01)

The *Citrullus lanatus* peel was removed using a stainless knife and an appropriate small piece was made. Approximate 500 gm of *Citrullus lanatus* peel were washed with distilled water and dried at 40ºC in a hot air oven and powder form was a maid and used future.

Preparation of bio-plastic film from watermelon peel (MR-02)

2gm Watermelon peel powder, 5gm corn starch, 3 ml glycerine, 3gm gelatine, and 1 gm citric acid combined in a 250-mL beaker. 100 mL of distilled water was added to the mixture. Stir the mixture for approximately 10–15 minutes. Following this, place the beaker on a heating mantle set to 100°C and manually stir for 1.5 hours. The reaction mixture concentrated up to 20% of the initial volume, poured onto a Teflon-coated glass plate spread, and allowed it to air-dry at ambient temperature. The drying process extended over a period of four days.

Weight of bio-plastic film: 9.49g

Preparation of bioplastic from watermelon peel with filtrated (MR-03)

2gm Watermelon peel powder, 5 gm corn starch, 3ml glycerine, 3gm gelatine, and 1gm citric acid combined in a 250-mL beaker, 100 mL of distilled water was added into the mixture. The reaction mixture is raised to 80–90 °C for 10-15 mins under stirring. Filtered this RM using a cotton cloth and heat the filtrate at 100 °C for 1–1.5 hours until the reaction mixture is concentrated up to 20% of the initial volume. Pour onto a Teflon-coated glass plate and allow it to air-dry at ambient temperature. The drying process extended over a period of four days.

Weight of bio-plastic film: 8.42g

Preparation of bioplastic from newspaper pulp and watermelon peel (MR-04)

5 gm Waste newspapers small pieces and 10 ml of water are added to them to obtain a lignocellulosic fibrous pulpy material that is stirred continuously to obtain what is known as paper sludge. This paper sludge was added to 100 ml of distilled water, 2 gm watermelon peel powder, 5 gm corn starch, 3 ml glycerine, 3gm gelatine, and 1 gm of citric acid. The mixture was heated at 130–140 °C, with manual stirring for 1–1.5 hours until the reaction mixture was concentrated up to 20% of the initial volume. Pour onto a Teflon-coated glass plate, spread, and allow it to air-dry at ambient temperature. The drying process extended over a period of four days.

Weight of bio-plastic film: 10.41g

Preparation of bioplastic from newspaper pulp and watermelon peel powder with filtrated (MR-05)

5 gm Waste newspapers small pieces and 10 ml of water are added to them to obtain a lignocellulosic fibrous pulpy material that is stirred continuously to obtain what is known as paper sludge. This paper sludge was added to 100 ml distilled water, 2 gm watermelon peel powder, 5 gm cornstarch, 3 ml glycerine, 3 gm gelatine, and 1 gm of citric acid. Stir the mixture for approximately 10–15 minutes. Following this, place the beaker on a heating mantle, maintaining a temperature of 80–90 °C for 10-15 mins. Filtered RM & heat the filtrate at 100 °C until the reaction mixture was concentrated up to 20% of the initial volume. Pour onto a Teflon-coated glass plate, spread, and allow it to air-dry at ambient temperature. The drying process extended over a period of four days.

Weight of bio-plastic film: 9.21 g

Preparation of bioplastic from newspaper pulp and watermelon peel powder (cost-cutting purpose) (MR-06)

10 gm of discarded newspaper was combined with 10 ml of water to create fibrous, pulpy material rich in

lignocellulose. This amalgamation was consistently stirred to form a sludge-like substance. The produced sludge was added into a mixture containing 100 ml distilled water, 5 gm watermelon peel powder, 3 ml glycerine, 1 gm vinegar (acetic acid), and 1gm of cornstarch. Stir the mixture for approximately 10–15 minutes. The mixture was heated at 130–140 °C, with manual stirring for 1–1.5 hours till RM attained 20% of the initial volume. Pour into a Teflon-coated glass plate and allow it to air-dry at ambient temperature. The drying process extended over a period of four days.

Weight of bio-plastic film: 15.71g

Table 1: Costing of the formulated Biodegradable polymer (as per wt. obtained)

Raw Material	Price/50 g	Quantity for Film (MR-02) & (MR-03)	Price For Film (MR-02) & (MR-03) (in INR)	Quantity for Film (MR-04) & (MR-05)	Price for Film (MR-04) &(MR-05) (in INR)	Quantity for Film (MR-06)	Price for Film (MR-06) (in INR)
Watermelon Peel Powder	37.5 Rs.	2g	1.5	2g	1.5	5g	3.75
Corn starch	2 Rs.	5g	0.2	5g	0.2	1g	0.004
Glycerine	40 Rs.	3g	2.4	3g	2.4	3g	2.4
Gelatine	18.5 Rs.	2g	0.74	-	-	-	-
Citric acid	2.7 Rs.	1g	0.054	-	-	-	-
Vinegar	2.5 Rs.	-	-	5g	0.25	1g	0.05
Waste News Paper	0.25 Rs.	-	-	5g	0.025	10g	0.05
TOTAL COST in (INR – Indian Rupees.)			4.894		4.375		6.164

Table 2: Comparison of final costs of synthesized biodegradable films

Film Sample	Wt. Obtained (gm)	Costing (in INR)	Costing per Kg (in INR)
MR-02	9.49	4.894	515
MR-03	8.42	4.894	581
MR-04	10.41	4.375	420
MR-05	9.21	4.375	475
MR-06	15.71	6.164	392

Costing of Film MR-06 is relatively more beneficial, 15.71 gm of film was obtained at the cost of 6.164 Rs. Film MR-06 is relatively less costly than other prepared films MR-02, MR-03, MR-04 & MR-05 in bulk. So, the designed experiment for cost-cutting purposes is successful.

RESULT AND DISCUSSION

Water Absorption Test

Two-centimetre square portions of a tiny piece of film were taken off. The cut-fill sample's starting weight was then calculated. The sample was then submerged for 24 hours in 50 ml of room-temperature water. After that, the sample was wiped off and removed from the water. Finally, the weight was noted. The following formula was used to determine the quantity of water absorbed. According to **Table 1**, the BP film MR-03 had the maximum water uptake.

$$\boldsymbol{WA}(\%) = \frac{\boldsymbol{Final\ weight\ (gm) - initial\ weight\ (gm)}}{\boldsymbol{initial\ weight\ (gm)}} \times \mathbf{100}$$

Table 3: Data for water absorption of BP films

Sample BP Film	Initial Weight BP film (gm)	Final Weight BP Film (gm)	Water Absorption Water Uptake (%)
MR-02	0.297	0.436	46.80
MR-03	0.228	0.388	70.17
MR-04	0.236	0.359	52.11
MR-05	0.219	0.330	50.68
MR-06	0.312	0.456	46.15

Biodegradability Test

Biodegradable plastics must decompose in certain situations, such as compost, soil or aquatic settings, by naturally occurring microorganisms such as bacteria, fungi and algae. Biodegradable tests were carried out in which BP film was passed in soil for several days and their degradation in soil was measured for environmentally friendly plastic uses. The BP film MR-05 degraded easily within a short period of time. As shown from the data given in **Table 4**.

Table 4: Data for Biodegradable test of BP films.

Sample BP film	Initial Weight BP film	Weight after 7 days (gm)	Weight after 10 days (gm)	Weight after 15 days (gm)
MR-02	0.992	0.841	0.737	0.638
MR-03	1.237	1.08	0.817	0.592
MR-04	1.729	1.193	0.872	0.412
MR-05	1.024	0.883	0.673	0.394
MR-06	1.682	1.369	1.277	0.876

Photodegradability test

A piece of all synthesized bioplastics is placed in sunlight to check the photodegradability study. After regular intervals of days, the weight of biodegradable plastic was checked. From this photodegradability study from looking at **Table 5** we can say that there are no significant changes in the weight of bioplastics so we can say that they are photo stable.

Table 5: Data for Photodegradability test of BP films.

Sample BP film	Initial weight gm	Weight After 7 days gm	Weight After 10 days gm	Weight After 15 days gm
MR-02	1.146	1.143	1.143	1.142
MR-03	1.126	1.124	1.124	1.124
MR-04	1.182	1.181	1.181	1.180
MR-05	1.177	1.176	1.176	1.176
MR-06	1.945	1.943	1.943	1.942

FTIR Characterization

FTIR Interpretation of *Citrullus Lanatus* Peels and bioplastic from watermelon peel (MR-01): Using the Spectrum 100 Bruker FTIR, the spectra were captured in 256 scans between 4000 and 400 cm^{-1}. The spectra of the watermelon peel powder and the watermelon Bp film were analyzed using FTIR and compared.

Due to the intricate vibrational stretching that naturally takes place in the carbohydrate structure, the peaks seen between 3100 and 3700 cm^{-1} in overlapping spectra correspond to the hydrogen-linked hydroxyl group (O-H). The shift of C-O stretch has been observed at a slightly lower side of the range as compared to the powder and bp film and a slight increase in the OH group. The sharp peak observed in the range 2850–2950 cm^{-1} is the C-H peak of the CH_2 group of the powder and bp film. It has been observed that there is a broad peak observed in the bp film. This is due to the moisture present in the film. Peaks around wavenumbers 1580 to 1700 cm^{-1} were seen in overlapping spectra and were attributed to the water's hydroxyl group deflection, which was particularly caused by hydroxyl groups bending the mode in water molecules. The other peak observed in both spectra was within the range of 1400–1450 cm^{-1} corresponding to O-H bend and peaks at 1350–1480 cm^{-1} were assigned to CH_2 bending vibration in the BP film, as shown in all figures of the IR data. Below are the overlapping IR data of *Citrullus Lanatus* Peels (Blue line (MR-01)) and bioplastic from watermelon peel (Red line (MR-02) & Pink Line (MR-03)) **Figure 1**.

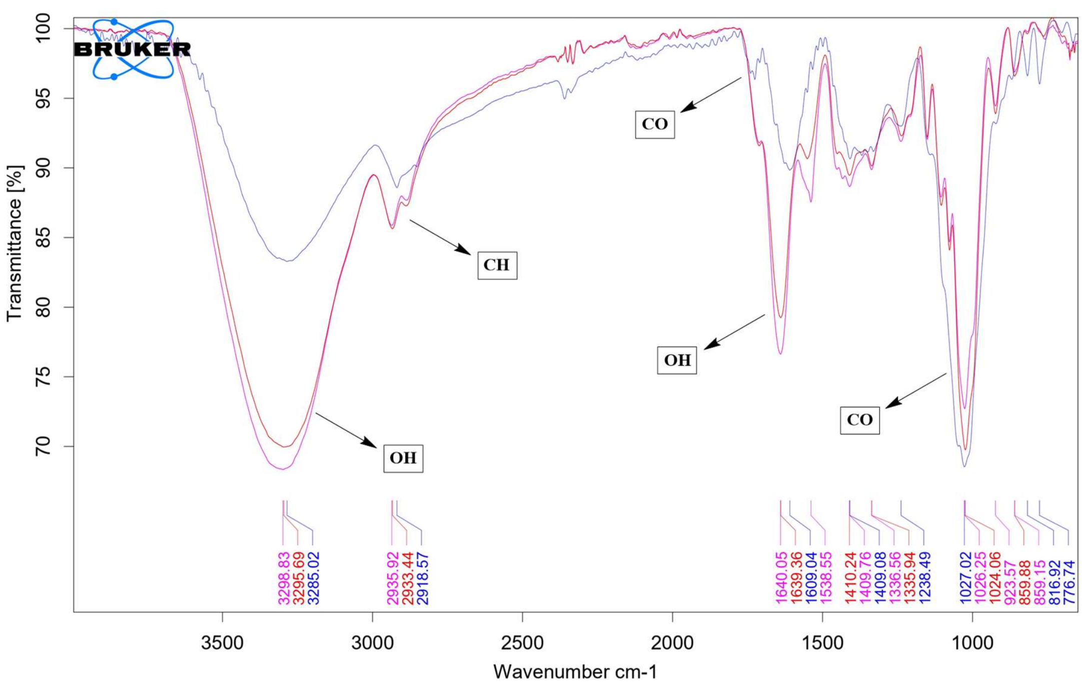

Figure 1: Overlapping ir data of *Citrullus Lanatus* Peels (Blue line) and bioplastic from watermelon peel (Red line (MR-02), Pink Line (MR-03))

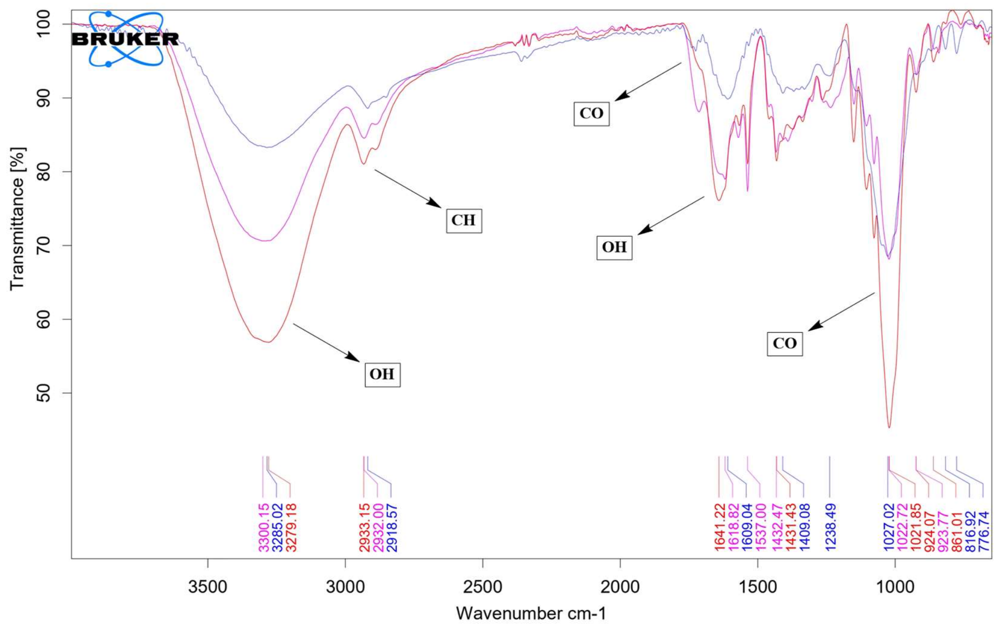

Figure 2: Overlapping ir data of *Citrullus Lanatus* Peels (Blue line (MR-01)) and bioplastic from newspaper pulp with watermelon peel powder (Red line (MR-04) & Pink Line (MR-05))

The Peaks observed are the same in the Overlapping IR data of *Citrullus Lanatus* Peels (Blue line (MR-01)) and bioplastic from newspaper pulp with watermelon peel powder (Red line (MR-04) & Pink Line (MR-05)) Figure 2. shows peaks ranging from 1400–1500 cm^{-1} corresponding to O-H bending, 1350–1480 cm^{-1} for the CH_2 bending vibration of bioplastic film, 1580–1700 cm^{-1} for the water hydroxy group, 3100–3700 cm^{-1} for the hydrogen-linked hydroxyl group (OH) stretching and 1000–1150 cm^{-1} for the C-O stretch of an anhydrous glucose ring. Hydrogen bonds interact specifically, causing the FTIR spectra to shift and widen.

Overlapping of IR data confirms the insertion of Citrullus Lanatus (Water melon) peel powder in the prepared bioplastic films as all functional groups present in IR spectra of Citrullus Lanatus peel powder are present in synthesized Bioplastic films.

X-ray diffraction (XRD)

In bioplastics, X-ray diffraction (XRD) is a commonly used technique to measure crystallinity. The polymers are never 100% crystalline. Figure (3) shows XRD spectra in which Black is MR-01(watermelon peel powder). Red is MR-04(bp made from Watermelon peel powder and newspaper with filtered) and blue is MR-05(bp made from Watermelon peel powder and newspaper) films. From the spectra, we can say that MR-01(watermelon peel powder) is an amorphous material as it does not show any sharp peaks. While MR-04 and MR-05 have a semi-crystalline nature. We can observe their peaks at 17.4°, 19.23°, 21.9°and 23.54° in MR-04 and MR-05. That is due to the Corn starch and Newspaper.

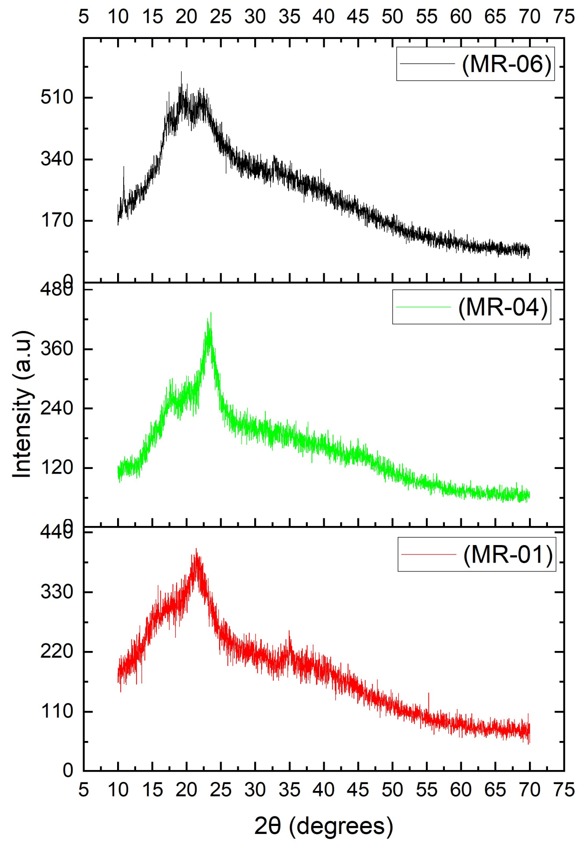

Figure (3): X-ray diffraction pattern of watermelon peel powder and bp films

Anti-Bacterial activity

The synthesized films were tested for their antibacterial activity against *Pseudomonas aeruginosa* (gm^{-ve}), *Escherichia. Coli* (gm^{-ve}) *Bacillus subtilis* (gm^{+ve}) and *staphylococcus aureus* (gm^{+ve}) using disc diffusion method at 100 ppm (10 mg/ml) concentration in DMSO (dimethyl sulfoxide) solvent. Using a disc diffusion technique described by the Kirby-Bauer method **[10]**. Penicillin-G was used as a standard drug. Each experiment was repeated twice. antibacterial test for all the bioplastic films, which is shown in **Table (6)**.

Table 6: Result of Antibacterial activity test

Antibacterial activity					
Name of sample	Conc. (µg/ml)	Microorganisms and zone of inhibition			
		Gram-positive bacteria		Gram-negative bacteria	
		S. aureus	*B. substilis*	*E. coli*	*P. aeruginosa*
MR-02	10	16mm	16mm	9mm	12mm
MR-03	10	15mm	14mm	10mm	13mm
MR-04	10	14mm	15mm	11mm	13.5mm
MR-05	10	17mm	10mm	12mm	10mm
Control (PenicillinG)	10	14mm	14mm	16mm	10mm

Looking at the result of the anti-bacterial activity test we can say that all of the films have good antibacterial activity towards *S. aureus* and *B. subsills* but MR-02 & MR-03 have better antibacterial activity against *S. aureus* and *B. subsills*.

CONCLUSIONS

The bioplastic film is formed successfully with watermelon peel and waste newspaper. The water absorption test shows all the bioplastic films are hydrophilic in nature. FTIR characterization of bioplastic film indicates different functional groups present in watermelon peel powder and the bioplastic film produced from it by identifying most of the peaks present in the IR spectrum. The overlapping of IR spectra clearly indicates the insertion of Citrullus Lanatus (Watermelon) peel powder to bioplastic films. XRD analysis tells us that some of the peaks show a crystalline nature and some show an amorphous nature as it proves bioplastics are never crystalline & comparison of XRD data also indicated the insertion of watermelon powder to the synthesized BP films as their spectras are similar. Synthesized bioplastic films show potent antibacterial activity against *S.aureus, B.Subtilis, E.Coli & P.aeruginosa*. What we seek are solutions that blend with nature, give value without damage and stay economically viable bioplastic. Envisioning a future resplendent with sustainability, the clarion cry is to boost the demand for bioplastics and usher in a new age of heightened manufacturing. With potential cost savings exceeding those of standard plastics, the appeal of biodegradability emerges victorious. Notably, Composition MR-04 has both cost-effectiveness and a noteworthy rate of commercial biodegradability. MR-06 composition with high amount of newspaper pulp is cost-effective & biodegradable. Emboldened by these prospects, our initiatives have delivered an assortment of biodegradable polymers harnessed from nature's richness, presenting a panorama of fascinating economic potential.

ACKNOWLEDGEMENTS

Gratitude is extended to Dr. Devanshu Patel, the President of Parul University, for his generous provision of essential facilities. A distinct distinction is also reserved for the Parul Institute of Pharmacy within Parul University for affording access to the FTIR facility.

REFERENCES

[1] Avinash Kumar, Vidhi Gupta, Suman Singh, Shashank Saini, Kirtiraj K. Gaikwad. Pine needles lignocellulosic ethylene scavenging paper impregnated with nanozeolite for active packaging applications. Ind. Crops Prod., 2021, vol. 170, p. 1-10. https://doi.org/10.1016/j.indcrop.2021.113752

[2] Ravindra V. Gadhave, Abhijit Das, Prakash A. Mahanwar, Pradeep T. Gadekar. Starch-based bioplastics: the future of sustainable packaging. Open Journal of Polymer Chemistry., 2018, vol. 8, p. 21-33. http://doi.org/10.4236/ojpchem.2018.82003

[3] Wafa A. Alshehri, Ashjan Khalel, Khaled Elbanna,Iqbal Ahmad and Hussein H. Abulreesh**.** Bio-plastic Films Production from Feather Waste Degradation by Keratinolytic Bacteria Bacillus cereus. J Pure Appl Microbiol., 2021, vol. 15(2), p. 681-688. https://doi.org/10.22207/JPAM.15.2.17

[4] Muhammad Mushtaq, Bushra Sultana, Haq Nawaz Bhatti, Muhammad Asghar. RSM based optimized enzyme-assisted extraction of antioxidant phenolics from underutilized watermelon (Citrullus lanatus Thunb.) rind. J. Food Sci. Technol., 2015, vol. 52, p. 5048–5056.

[5] Reem Nadher Ismael, Yasser Fakri Mustafa, Harith Khalid Al-Qazaz. Citrullus lanatus, a Potential Source of Medicinal Products: A Review. Journal of Medical and Chemical Sciences., 2022, vol. 5(4), p. 607-618. https://doi.org/10.26655/JMCHEMSCI.2022.4.16

[6] Pavan Kumar, Nitin Mehta, O. P. Malav, Manish Kumar Chatli, Manjeet Rathour, Akhilesh Kumar Verma. Antioxidant and antimicrobial efficacy of watermelon rind extract (WMRE) in aerobically packaged pork patties stored under refrigeration temperature (4 ± 1 °C). J. Food Processing Preserv., 2018, vol. 42, p. 1-12. https://doi.org/10.1111/jfpp.13757

[7] Neglo David, Tettey Clement Okraku, Essuman Edward Ken, Kortei Nii Korley, Boakye Adjoa Agyemang, Hunkpe Gaston, Amarh Flora, Kwashie Pius, Devi Waikhom Sayanika Comparative antioxidant and antimicrobial activities of the peels, rind. pulp and seeds of watermelon (Citrullus lanatus) fruit Scientific African., 2021, vol. 11, p. 1-8. http://doi.org/10.1016/j.sciaf.2020.e00582

[8] Molka Ben Romdhane, Anissa Haddar, Imen Ghazala, Khawla Ben Jeddou, Claire Boisset Helbert, Semia Ellouz-Chaabouni. Optimization of polysaccharides extraction from watermelon rinds: Structure, functional and biological activities. Food Chem., 2017, vol. 216, p. 355–364. https://doi.org/ 10.1016/j.foodchem.2016.08.056

[9] Harsh Kumar, Kanchan Bhardwaj, Ruchi Sharma, Eugenie Nepovimova, Kamil Kuča, Daljeet Singh Dhanjal, Rachna Verma, Prerna Bhardwaj,

Somesh Sharma, and Dinesh Kumar. Fruit and Vegetable peels: Utilization of high value horticultural waste in novel industrial applications. Molecules., 2020, vol. 25(12): 2812. http://doi.org/ 10.3390/molecules25122812

[10] A W Bauer, W M Kirby, J C Sherris, M Turck. Antibiotic susceptibility testing by a standardized single disk method. Am J Clin Path., 1996, Vol. 45(4): p. 493-496. https://doi.org/10.1093/ajcp/45.4_ts.493