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EXTRACTION, CHARACTERIZATION AND DEVELOPMENT OF CELLULOSE BASED BIO-FILM FROM OIL INDUSTRY WASTE AND AGRICULTURAL PLANTS

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Abstract

Bio polymers are polymerized high molecular constituents from natural substances from the natural substances by the chemical or biological reactions. The most common biopolymers are polynucleotides, polyamides, polysaccharides, polyoxoesters, polythioesters, polyanhydrides, polyisoprenoides polyphenols, and their derivatives. Among the polysaccharides the starch, chitin, and cellulose are widely used biopolymers from the plant source. In this current study cellulose was extracted from oil industry waste such as sesame oil cake (S*esamum indicum)*, peanut oil cake *(Archishypoogaea)*, andcoconut oil cake (*Cocus nucifera)* and the plant samples ofpalm (*palmae)* and taro (*Colocasia esculenta)*. The extracted crude cellulose was estimated by dry weight, and UV-spectrophotometric method. The functional properties are characterized using Fourier Transform Infrared (FTIR) analysis. Three different sample of palm leaves, sesame oil cake and coconut oil cake yielded the better quantity of 18.14 g (w/w), 10.78g and 9.80 g in dry weight analysis than the other samples. Hence, the spectrophotometric quantification resulted the cellulose content of 0.25g/100g in palm leaves, 0.17 g/100g in sesame and coconut oil cake samples. The FTIR analysis also confirms that the structural properties of cellulose in these samples. Hence, the developed biodegradable film from palm leaves, sesame have the better morphology in SEM analysis. Therefore, the study results conclude the palm leaves and sesame oil cake were the suitable raw material for extraction and development of cellulose based biodegradable film.

Keywords: Cellulose, Agricultural Plant, Oil Industry waste, Biodegradable, FTIR

Introduction

Cellulose had versatile application in many industries among them food, pharmaceutical and cosmetic industries are highly demanded. Cellulose is the organic structural component of plants, natural long chain polymer, composed of linear segments (1 → 4)-linked b -D-glucopyranosyl units, water-insoluble and exhibited in crystalline structure(Guadalupe et al., 2009). The derivatives of methylcellulose, carboxymethyl cellulose, hydroxypropyl cellulose, hydroxypropyl methylcellulose, and microcrystalline cellulose have unique physical, chemical and colloidal properties, and an ability to form packaging films(Kester and Fennema, 1986).Packaging film plays animportant role in the food as a protecting barrier, by controlling the gas and vapor exchanges in the storage atmosphereand extending their food shelf-life of the product through evade the food safety issues like food-borne diseases, and food contamination(Guillard, et al.,2018). The packaging film should contain substances that cooperate with food regulations, and shouldalso be economical, relevant, and eco-friendly. Moreover, it preserves the appearance, mechanical and nutritional properties and protect volatilecompounds in the foods(Yildirim-Yalcin, et al.,2022). Among the various packaging material, the plastic was the major contributor among the countries, especially in the middle and the low-income due to the flexibility and economic feasibility(Yanti, et al., 2021). The plasticin the natural environment (air, soil, and water) affects their structure, permeability, C and N cycles, greenhouse gases, nutrient transfer, microbes, plants, animals and humans. Moreover, the impact of plastics on the global environment and their existence of micro-plastic (up to 25 particles/L) after the waste management methods demands the need of biodegradable packaging materials for the sustainable food packaging(Ahamed et al., 2021).The cellulose from the renewable sources are mainly used in biodegradable packaging(Vroman et al., 2009). Devi et al. (2021) who have stated that the application of various composites of cellulose on fruits and vegetables packaging extend the shelf life with the quality. Moreover, the odorless, and tasteless properties of cellulose are extending their application as anedible film for many fruits. In order to increase the cellulose-based bio-films, the oil industry waste and agricultural plants were selected for the extraction of cellulose.

MaterialsandMethods

Collection of samples

The waste samples from oil industry namely sesame oil cake (*Sesamum indicum*), peanut oil cake*(Archishypoogaea*), coconut oil cake (*Cocus nucifera*) was collected from Uzhavan oil industries which werelocatedinCoimbatore (Figure 1)Tamilnadu,India.Theplantleaves of palm(*Palmae*) andtaro(*Colocasia esculenta*) (TL) were collected in agricultural fields of coimbatore, Tamil Nadu,Indiainthemonthof January2023.

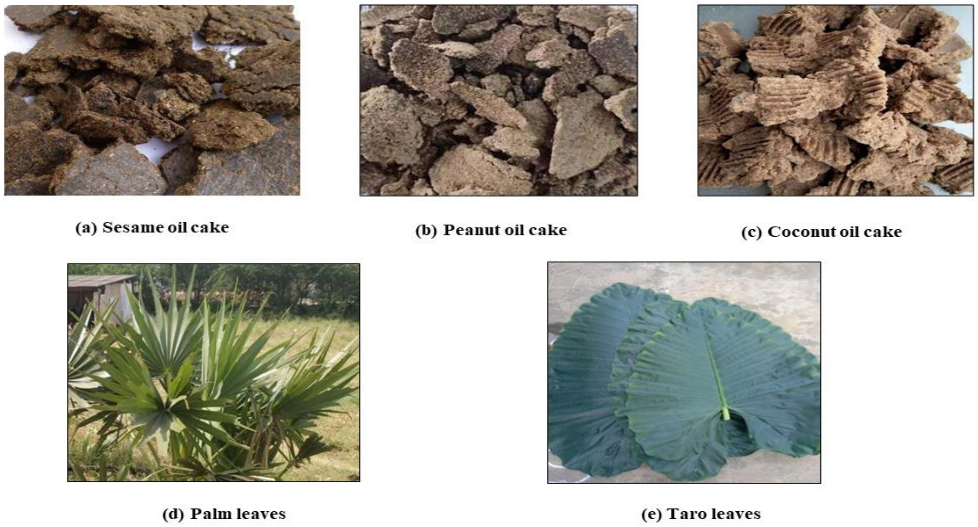

Figure 1*: Selected samples for the extraction of cellulose (a) Sesameoilcake- Sesamum indicum; (b) Peanutoilcake- Archishypoogaea; and (c) Coconutoilcake- Cocus nucifera from oil industry waste (d) Palm leaves- Palmae and (e) Taroleaves-Colocasia esculenta*

Extractionofcellulosefrom oilindustrywastesandplant leaves

Samplepreparation

The collected plant sample of palm leaves and taro leaveswere washed remove dust and adhered soilsparticles. After washing, leaves were cut into pieces (1-4 cm) and dried in a hot airovenat60°Cforthe period of 48h.Dried plant leaves and oil industry wastes werehomogenized usingmortarandpestle togetfinepowder.

Alkalinepretreatmentmethod

Thegroundedsamplesof30gweredissolvedin450mLoftoluene:ethanol(2:1)andleftfor shakingin a temperature-controlled orbital shaker(RemiRIS-24,India)at 150 rpm and 25 °C for 20h(Romruenet.al.,2022).After shaking the sample was filtered through (Whatman No. 1) filter paper. Thereafter the solvent removedfibers,werewashedfor twiceusingabsoluteethanolanddriedat100°Cfor1h.

Bleachingprocess

Driedfibrousmatterofthesampleswasbleachedaccordingtotheprocedurebyusing 1.4% (w/v) sodium chloride (NaCl)Romruen et al., (2022). After bleaching the oil industry waste and plant leaves wereadjusted to pH of 4 by adding 5 % (v/v) acetic acid solution followed by heating at 70 °C with continuousstirring using an overhead stirrer (Remi 1 MLH, India) at 500 rpm for 5 h. From the heated sample fibers are filtered and washed with sodium hydroxide(NaOH-4%(v/v)untilthepHadjustmentof7,washed samples were dried at 100 °C for 16 h. The dried fibers were soaked in 600 mL of 5% (w/v)potassiumhydroxide(KOH)andstirringwascontinuedin 500rpmat ambienttemperaturefor24hpriortoheatingat90°Cfor2h, for theextractionofcellulose.TheKOHtreatedsampleswerewashedwith distilled water then the pH was adjusted to neutral by using 5 % (v/v) acetic acid solution.Followed by pH adjustment for the extraction of cellulose the samples were dried using hot-air ovenat100°Cfor1h.

Estimationofcellulose

The0.5 g of sampleswere added 3.0 ml Acetic/Nitric reagent. The acidified tubesplacedin water bath at 100 °C for 30 min. After the heating process the tubes were cooled.The cooled tubes were centrifuged at 1200 rpm for 15-20 min. The collectedresiduefrom the centrifugation were washed thoroughly. Thereafter, 10 ml of 67 % H_2SO_4were added into residue and allow for 1 h. The diluted sample of 1 ml added in series of volumes i.e., 0.4, 0.6, 0.8, 1.0 and 2.0 ml.Finally, the10mlofanthronereagentwere added into all the test and blank tubesand it boiled up to 10 min. After incubation the absorbance was measured at 630nm in UV spectrophotometer.

Fouriertransforminfrared spectroscopy(FTIR)

FTIRanalysis were carried out using aPerkin Elmer L1600300 Spectrum Two LiTa.The powdered samples were pelletized with KBr and spectrum from the wavelength of4000 to 400 cm^{-1}were recorded.

Film preparation

Theextractedcellulosefrom the samples dissolvedinwaterattheratioof1:8and blended sample until it became slurry. The slurry was used as the essential ingredient for making the packaging film.In that cellulose slurry the additive compounds of carboxymethylcellulose(0.5%)andglycerol(0.5%) were added.Thereafter,thecompounds were mixed samples and kept on hot plate at 80 °C for 10 min up to the even mixing with the additive compounds. Then the mixed sample was poured in the glass platesdriedat40°Cin hotairovenfor24h.

Scanningelectronmicroscopy(SEM)

SEManalysis was performed to analyze the surfacemorphology characteristics of the packaging film layer and characteristic of cellulose.Thefilms during analysis coatedon the goldandexaminedatanacceleratedvoltageof5kVthe specimen were captured using CAREL ZEISS, EVO 18.

ResultsandDiscussion

Extractionand quantification ofcellulose

In thispresent study30 g of each sample were yielded the cellulose after alkali treatment. Among the five samples (oil cake of sesame, peanut,coconut andleaves ofpalm andtaro) revealed the high concentration of cellulose in dry weight basissuch as 18.14 g (w/w), followed by sesame, coconut, taro leaves and peanut resulted the cellulose as 10.78g, 9.80 g, 8.46 g,and 6.53 g(w/w) respectively (Table 1). The resulted high yield in plant sampleof palm leaves was occurred from the highconcentrationofcellulosefibers, andtheir tight arrangement withinthecellwalls(Cárdenas et.al., 2016).The arrangementexhibited the highest yield from the 30 g of sample. In sesame sample the main component are cellulose, and hemicellulose in smaller amount.Zhang et al. (2022) who have reported the higher concentration in non-fibrous cellulose in sesame, the non-fibrous celluloseeasily breakdown during the alkali digestion.The effectresulted the concentration of 10.78g cellulose in the current study. Hence, the existed lignin and hemicellulose in the coconut weredissolute during chemical treatment. The fibrils cellulose in coconut are rearranged interfibrillar regions during the extraction, the effect may decrease the quantity of cellulose yield in extraction (kalla et al., 2022).Generally cellulose content of peanut was comparably lower than the other plants, because theiroccurrence reported in seed coat. The processing method involved in the peanut oil extractionaffectsthecelluloseyield(Ravindranetal.,2019).

Quantification ofcellulosebyUVspectrophotometricmethod

Amongtheanalyzedsamplesthepalm leaves andsesame oil cakerevealedtheconcentrationof0.25g/100g and 0.17 g/100 g respectively in Table 1.The highest concentration in the palm samples was due to the α-cellulose, and it widely resistant todegradation (Kudre et.al., 2012). Thesimilar concentration 0.17 g/100g of cellulose in sesame was reported in earlier study along with other impurities(Jayasingheet.al.,2012). Hence, the 0.17 g/100 g in peanut samples enriched with β-cellulose and this form of cellulose is more easily broken down and digested during the process(Huang et.al., 2015). The samples of taro leaves had the presence of α-cellulose in smaller amounts which resulted the quantity of 0.15 g/100 g of cellulose.

Table 1: Yield and Quantification of extracted cellulose from various samples

Name of the samples				
Sesameoilcake (*Sesamum indicum*)	**Coconutoilcake** (*Cocus nucifera*)	**Peanutoilcake** (*Archishypoogaea*)	**Palm leaves** (*Palmae*)	**Taroleaves** (*Colocasia*)
Yield of cellulose on dry weight basis (w/w)				
10.78±0.02	**9.85±0.03**	**6.53±0.01**	**18.14±0.01**	**8.46±0.04**
Spectrophotometric quantification ofcellulose (mg/ 100 g)				
0.17±0.02	**0.17±0.02**	**0.17±0.01**	**0.25±0.02**	**0.15±0.02**

Note: Extractedpowderfromoilindustrywastesandplantleavesbyalkalineandbleachingprocess. Palm leaves (*Palmae*), Sesameoilcake (*Sesamum indicum*), Coconutoilcake (*Cocus nucifera*),Taroleaves (*Colocasia*), andPeanutoilcake (*Archishypoogaea*)

FourierTransformInfraredSpectroscopy(FTIR)

The FTIR spectrum from palm leaves, sesame oil cake, coconut oil cake, taro leaves and peanut oil cake were depicted in Figure 4, and it typically analyzed for the characteristic peaks associated with cellulose's molecular structure Table 2. The O-H stretching in FTIR usually appears around 3200-3600 cm^{-1}, representing the hydroxyl groups (O-H) of cellulose (Reddy et al., 2016) and the peaks observed in all the analyzed samples. C-H stretching between 2900-3000 cm^{-1}, representing the stretching vibrations of C-H bonds in the cellulose structurewhich was another important structural property of cellulose (Zhang et al., 2021). The similar C-H peak was foundin palm leaves, sesame, coconut,and peanut sample at the peak of 2900 cm^{-1}. The sharp and medium band of 2026 cm^{-1} related to alkene groups (C-C) of cellulose and this peak was observed in palm leaves, coconut and taro leaves. The trans double peak around 1645-1660 cm^{-1}, indicating the presence of unsaturated fatty acids in the oil sample mainly sesame oil cake, coconut oil cake, peanut oil cake, and palm leave samples. Typically, peak seen at around 1630 cm^{-1}, indicating the presence of acetyl groups in hemicellulose or other compounds associated with the cellulose.The β- glycosidic linkages (C-O-C) asymmetrical stretching of cellulose which were related to the structural properties of cellulose are exhibited from 1000-1150 cm^{-1} (Zhang et al., 2021). This glycosidic bond was reported in the palm leaves, sesame, coconut and peanut. Guadalupe et al. (2009) who have reported the development of cellulose film the structural properties are more important. And these structural properties of cellulose mainly reported at sharp peak in FTIR spectra for the sample of palm leaves at (1113 cm^{-1}), and sesame (1100 cm^{-1})sample.From the FTIR spectrum it is suggested that sesame oil cake and palm leaves constitute an interesting and promising alternative as cellulose source and derivatives. Therefore, sesame oil cake and palm leaves sample are suitable for biofilm formation and can be used as packaging material.

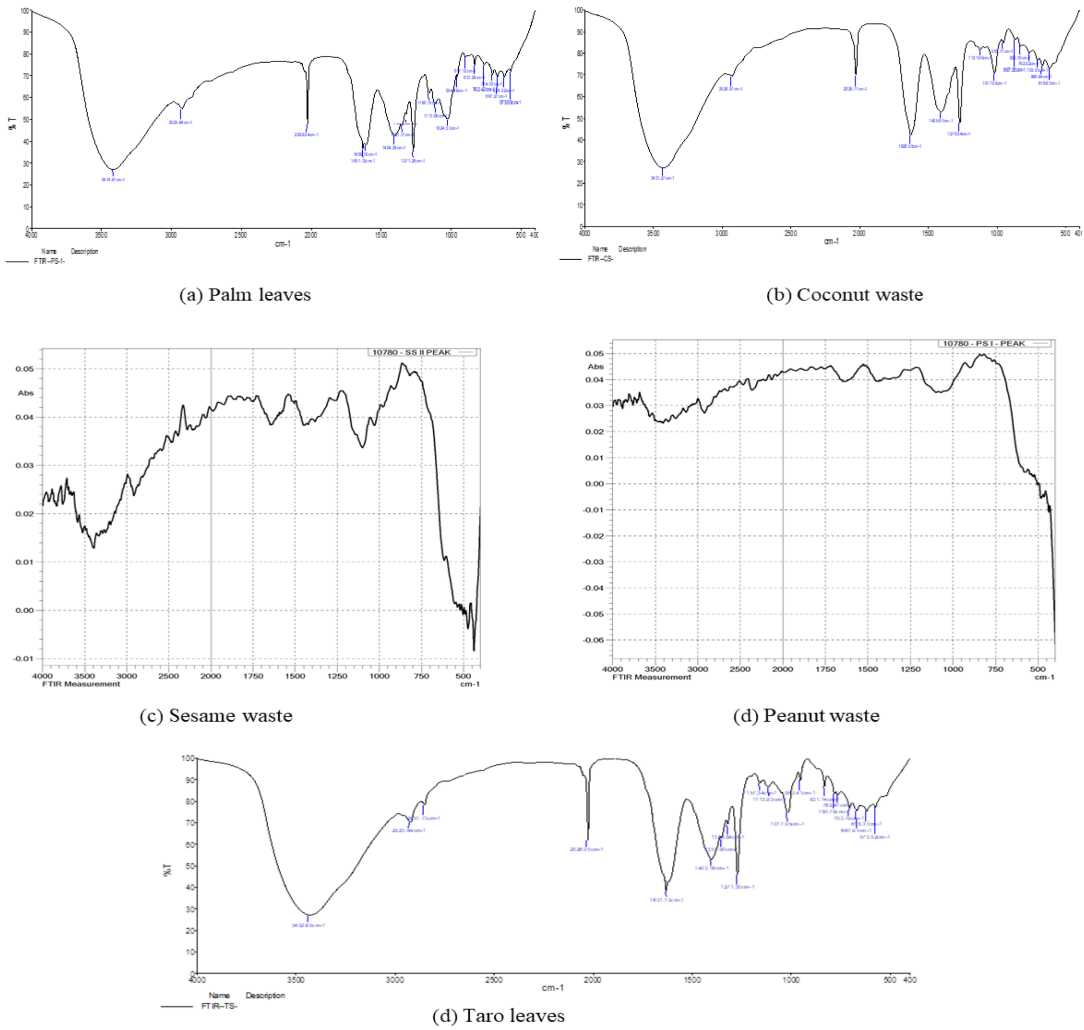

Figure 2: FTIR spectra after cellulose extraction from various samples, (a) Palm leaves- Palmae, (b) Coconutoilcake- Cocus nucifera (c) Sesameoilcake- Sesamum indicum, (d) Peanutoilcake- Archishypoogaea (e) Taroleaves-Colocasia

Table 2 Major band assignments for the IR spectra of Palm leaves- *Palmae,* (b) Coconutoilcake-*Cocus nucifera* (c) Sesameoilcake- *Sesamum indicum*, (d)Peanutoilcake-

Archishypoogaea*(e)Taroleaves-*Colocasia,

Palm leaf	**Coconut oil cake**	**Sesame oil cake**	**Peanut oil cake**	**Taro leaves**	**Attribution**
Wavenumber (cm^{-1})					
3414	3431	3400	3400	3432	Symmetric and asymmetric vibration of O-H
2925	2928	2900	2900	-	C-H symmetrical stretching
2026	2026	-	-	2029	(C-C)
1609	1628	1600	1600	_	Tans double Bonds
1631	1637	-	-	-	Acetyl group
1404	-	-	_	1403	CH2 bending ($1480\ cm^{-1}$)
1271	1270	-	-	1271	C-O phenolic OH
1351		-	-	-	=C-O-C Symmetric stretching
1160	-	-	-	-	In plane bending of aromatic C-H
1113	-	1100	1100	-	=C-O-C symmetric stretching
1024	1017	-	-	1017	=C-O-C symmetric stretching

Scanningelectronmicroscopy(SEM)results

SEM analysis was carried out to determine the surfacemorphologyand cross-sectional views ofthe developed films at 100 X and 2 X magnification in Figure 3.TheSEMmicrographs illustrated the different morphologyof cellulose for the analyzed samples. It was noted that film developed from the palm leaves showed thethread-like structures of cellulose fibers, with dark in appearance. The appearance indicates the cellulose in the palm leaves are usually appear as long and slender in structures and interconnected with other components such as lignin and hemicellulose (Pongchaiphol et al., 2022). Thus provided the rough morphology of the developed film out of palm leaves. The sesame waste revealed smooth or compact surfaces than the other film. The SEM of sesame showed the cellulose fibers on the surface exhibit a certain length and thickness(Figure 3) the arrangement and density provide a smooth structure(Romruen et al., 2022).Hence, the surface morphology of coconut waste shows the irregular arrangementof cellulose on the surface and their lower magnification shows the amorphous cellulose (crystal) structure, but their occurrence provided rough surface. The image of coconut film confirms less quantity of cellulose in the coconut peduncles. The study results in coconut oil waste were closely related with the earlier study(Nagarajan et al., 2019). Who have reported the least concentration of α-cellulose which were not produced the compact surface in the developed film. The difference shows the palm leaves and sesame film expected to have better morphology for the film development.

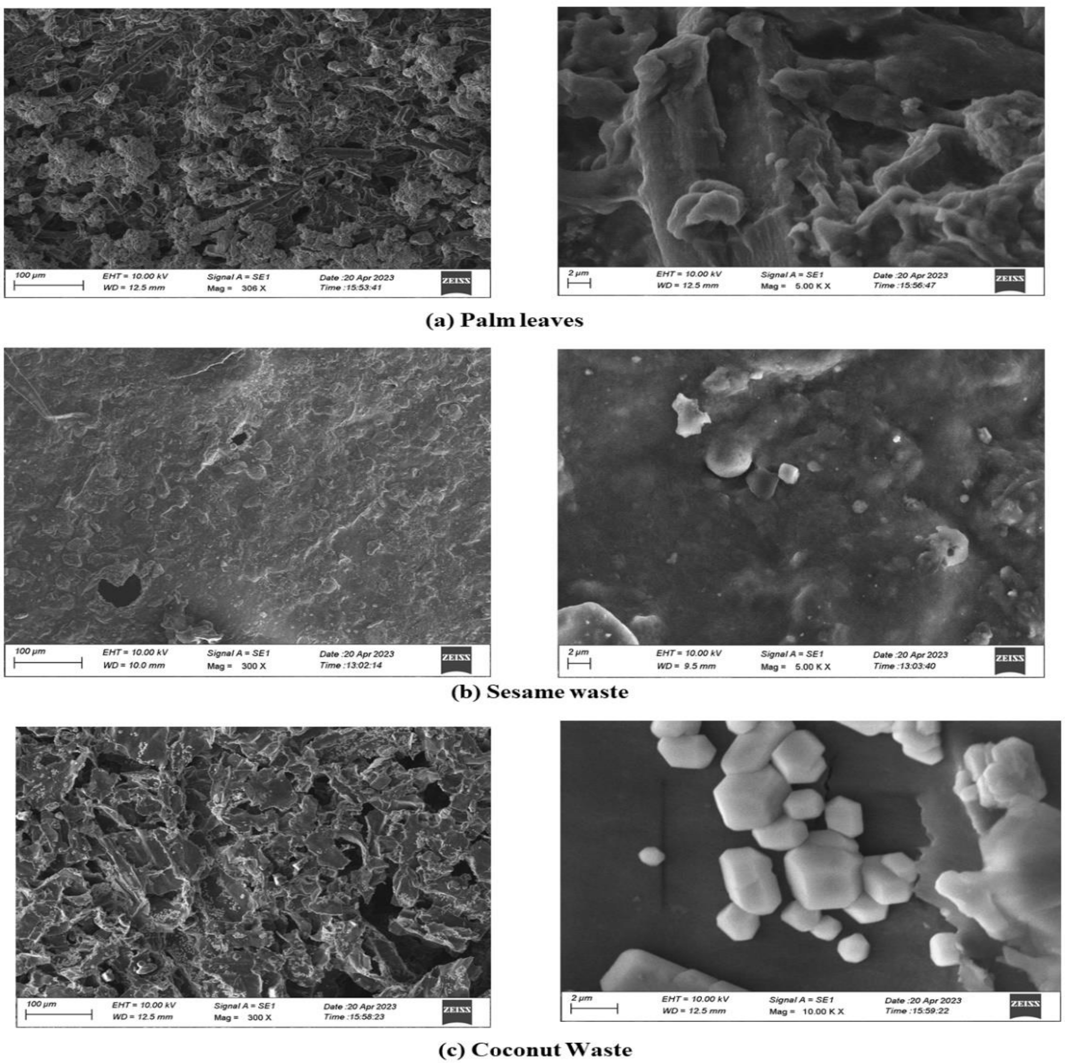

***Figure 3*: Scanning Electron Micrographs of developed cellulose film with the alkaline and bleaching processing for the industry waste sample and agricultural plants. (a) Palm leaves- *Palmae*, (b) Sesamewsate- *Sesamum indicum*, (c) Coconutwaste-*Cocus nucifera*.**

Conclusion

The natural polymer of cellulose are successfully extracted from oil industry waste and natural plants after alkali and bleaching process. Three different sample of palm leaves, sesame oil cake and coconut oil cake yielded the better quantity of 18.14 g (w/w), 10.78g and 9.80 g than the other samples. The quantified cellulose of 0.18g/100g in palm leaves and sesame oil cake, 0.17 g/100g in and coconut oil waste by spectrophotometric are confirmed the existence of cellulose in these sample. The observed result in the current study could be a useful for selection of raw material for the extraction of cellulose. The developed biodegradable film from palm leaves, sesame oil cake has the better morphology than the coconut-based film. The smooth appearance showing the gelling characteristics of cellulose in sesame oil cake is the suitable raw material for the extraction of cellulose and development of biodegradable film. The other impurities in the palm leaves could be removed and thus can be used as a effective film for food application.

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