EXTRACTION AND CHARACTERISATION OF CELLULOSE FROM RED SEAWEEDS OF *Hypnea musciformis* AND *Sarconema filliforme*

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The study shows a comparison of cellulose extracted from two species of red seaweeds, namely *Hypnea musciformis* and *Sarconema filliforme*. The celluloses were characterized by FTIR, XRD, TGA and SEM analyses. The studies show similarities in the characteristics of the celluloses extracted from *H. musciformis* and *S. filliforme*. FTIR analysis confirms the presence of O-H and C-H bonds in the celluloses of both species, while the XRD patterns of celluloses confirm their crystallinity, with a maximum peak at 22°. The thermal stability of the celluloses from *H. musciformis* and *S. filliforme* was observed in the range of 250 to 350 °C. The morphological structure of the celluloses was studied using SEM and both celluloses showed smooth pore-free surface.

Keywords: marine, cellulose, biopolymer, seaweed, characterization

INTRODUCTION

The concept of biomaterials has been frequently advanced over the recent years, and significant progress has been reached in the design and utilization of safe and functional materials.[1,2] Biomaterials are natural, synthetic or semi-synthetic substances intended to be inserted into biological environments.[3,4] Natural and synthetic biomaterials are often combined targeting specific applications, natural biomaterials being mostly polysaccharide-based, including cellulose, chitin, alginate, starch, *etc*., or protein-based, *i.e.* albumin, collagen, gelatin, *etc*., derived from plants and animals. They have certain advantages over synthetic polymers, such as biocompatibility, biodegradability, extensive availability and unique biological activities.[5] Therefore, biomaterials have found successful clinical applications in reconstructive plastic surgery, orthopedic surgery, cardiac surgery, dentistry and other biomedical fields.[6-9]

In recent years, marine polysaccharide-based nanomaterials have attracted increasing attention, particularly, in biomedical and chemical research, due to their good biocompatibility, biodegradability, non-toxicity, low cost and abundance.[10] A variety of bioactive compounds, with various biological activities, are extracted from marine algae, which are known as a rich source of sulfated polysaccharides, including fucoidan, alginate, carrageenan, agarose, and ulvan.[10] Marine polysaccharides have many advantageous biological activities, having anticoagulant, antioxidant, anti-inflammatory, antiproliferative and immunomodulating effects.[11-13] To benefit from their properties, nowadays, marine polysaccharides can be turned into various forms – nanoparticles, nanofibers, microparticles, gels, beads and sponges.[14]

Cellulose is a linear polysaccharide composed of β-1,4 linked D-glucose units. Cellulose is known for its particular properties, such as high mechanical strength, chemical stability, biodegradability and a multitude of chemical derivatives.[15,16,26,27] Moreover, it is biodegradable and has a lower environmental impact compared to synthetic polymers obtained from fossil sources. Cellulose is insoluble in water and in most common solvents, because of its strong inter- and intramolecular H-bonding between its individual chain units. In spite of its poor

solubility, its good mechanical properties and wide availability in nature have motivated extensive research interest,[17] which led to many applications of cellulose in very diverse areas, including waste treatment, oil recovery, paper manufacturing, textile finishing, food additives, pharmaceuticals *etc*.[18] Its vast industrial applications include coatings, cigarette filters, textile fibers, filtration membranes, composites, laminates, drug delivery systems *etc*.[19-21]

Natural cellulose fibres have been derived from a variety of sources, including cotton stalks, rice and wheat straw, cornstalks and husks, which have shown qualities similar to those of cellulose obtained from traditional sources, such as cotton, jute, and linen.[22-25] Terrestrial plants are rich sources of cellulose, but it is intertwined with lignin, hemicelluloses and pectin in their structure, which demands pretreatments for their removal, making the extraction process expensive. Therefore, seaweeds have been also investigated as raw materials for the extraction of cellulose.[28] Nowadays, seaweeds are gaining prominence as an alternative renewable feedstock for fuels and chemicals, due to their high carbohydrate content, high productivity and widespread distribution across diverse geo-climatic regions.[29]

The current study investigates the possibility of extracting cellulose fibers from two species of red seaweeds – *Sarconema filliforme* and *Hypnea musciformis*. The red seaweeds were collected from the coast of Thondi, which is located in the Palk Bay region on the south-eastern coast of India. Palk Bay is a semi-enclosed shallow water body. Because the highest water depth is 13 meters, there are a lot of seaweeds in this area. Seaweeds are widely distributed on rocks, in lower intertidal and subtidal regions. Seaweeds are macroscopic, multicellular marine algae, plant-like organisms that generally live attached to rock or hard substrata in coastal areas. There may be several advantages in extracting cellulose from seaweeds, including their carbohydrate-rich content, high productivity, higher biomass per unit area, zero competition with agricultural land, no use of agricultural inputs, such as pesticides, fertilizers and water, and potential large-scale production.[30,31] Thus, this study aimed to investigate the viability of extracting cellulose from *Sarconema filliforme* and *Hypnea musciformis*. Structural characterization of obtained celluloses was performed by Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), thermogravimetric analysis (TGA). The structural topography of the celluloses was studied using scanning electron microscopy (SEM).

EXPERIMENTAL

Collection of seaweeds

The genus *Hypnea* is identified by a prostrate thallus, cylindrical to flattened, with a membranous to cartilaginous consistency. Thallus organization is erect, uniaxial and pseudoparenchymatous, with a single or more main axes, apical cell development, and apex shapes ranging from straight to curved, bifurcated, and tendril-like. The length of the thallus usually varies in the range of 0.5 to 50 cm, depending on the species habitat.[32] The colour of *Hypnea* is variable, with yellowish, greenish, pink, red, vinaceous, brown or blackish specimens. The genus *Sarconema* is characterized by an erect thallus, dark red or brown in colour. The subtropical species can grow to a length of 10-30 cm.[33]

Hypnea musciformis (Fig. 1) and *Sarconema filliforme* (Fig. 2) were obtained from the local fishermen, from the Thondi Coast (9º 45' N, 79º 04' E). The collected seaweeds were naturally dried for seven days. The dried seaweeds were weighed and ground before further use.

Extraction of cellulose

The extraction of cellulose was performed as per the methodology of Szymanska-Chargot (2017).[32] The extraction method proposed here involved four chemical treatment steps; each step was followed by rinsing with distilled water until a neutral pH was reached. The samples (4 g of powder sample) were refluxed in 40 mL of sodium hypochlorite (NaClO) solution (3%, v:v) at 100 °C for 10 min. After washing with distilled water, the procedure was repeated. Demineralization was done by refluxing the samples in 20 mL of 1 M HCl at 75 °C for 15 min. Then, the samples were refluxed in 20 mL of 1 M NaOH (sodium hydroxide) solution at 100 °C for 20 minutes to get rid of any protein residues. Finally, the extracts were filtered off and placed in an oven at 60 °C for five days. The obtained dry mass was cellulose and was subjected to further characterization. The cellulose obtained was washed multiple times with running water until a neutral pH was reached.

The results obtained for the amount of extracted cellulose, in terms of yield percentage, from the samples (n=30) were subjected to Students' t-test, to test the significance of variations within samples.

Characterization methods

Fourier transform infrared spectroscopy (FTIR)

Potassium bromide (KBr) supported cellulose samples were subjected to FTIR analysis over the frequency range between 4000-400 cm^{-1} at a resolution

of 4 cm^{-1}, employing a Perkins-Elmer spectrometer (Spectrum RX I, MA, USA).

X-ray powder diffractometry (XRD)

X-ray diffraction analysis (XRD) was conducted for determining the crystallinity of the extracted cellulose. An X'Pert PRO PAN Analytical (the Netherlands) instrument was operated at 40 kV and 30 mA with Cu kα radiation (1.5406A0).

Figure 1: Photograph of *Sarconema filiforme*

Scanning electron microscopy (SEM)

For analysis, the sample was placed on carbon tape and then spin-coated with a gold layer. A TESCAN SEM (Oxford) was used to record the scanning electron micrographs obtained at various magnifications.

RESULTS AND DISCUSSION

Extraction of cellulose

Cellulose was extracted from *Sarconema filliforme* and *Hypnea musciformis* samples with an average weight of 60 g and 30 g, respectively. The amount of cellulose extracted from *Sarconema filliforme* (60 g) and *Hypnea musciformis* (30 g) ground powder was 0.1826 ± 0.02 mg (0.3%) and 0.520 ± 0.01 mg (1.7%), respectively. The variations observed in the yield percentage of cellulose within the samples were statistically significant ($t<0.01$). The percentage of cellulose yield obtained in the present study from the two species of seaweeds is lower than that reported earlier from *U. fasciata*.[30] Considering the available literature on the extraction of cellulose from various species of seaweeds, it was noted that the highest yield percentage of crude cellulose was obtained from *C. sinuosa* (11.70% ± 0.92%), followed by *U. fasciata* (10.04% ± 0.82%), *U. linza* (5.78% ± 0.05%), *P. pavonica* (3.77% ± 0.32%), and *U. lactuca* (2.2%).[34-38] In contrast, much lower yields were obtained from *J. rubens* (1.38% ± 0.14%), followed by *A. rigida* (1.88% ± 0.12%), which can be explained by the calcareous nature of these species. The lowest yield of cellulose reported so far in the literature was obtained from *S. scinaioides*, which gave 0.3% cellulose, as shown by Siddhantha *et al*.[33]

Thermogravimetric analysis (TGA)

A Mettler Toledo TGA2 was utilized to ascertain the thermal behavior of the cellulose samples. The samples weighed around 5 mg. The thermal resistance of the materials was evaluated at temperatures ranging from 0 to 600 °C, at a nitrogen flow rate of 50 mL/min, and a heating rate of 10 °C/min.

Figure 2: Photograph of *Hypnea musciformis*

Consistent with our results, it appears that calcareous species of Nemaliales (Rhodophyta) produce the lowest cellulose content, because of their very high $CaCO_3$ content in the cell wall matrices.[34-38] Apart from the presence of carbohydrates, seaweeds are known to contain high amounts of calcium, magnesium, sodium, potassium and iron, which need to be neutralized in order to extract the cellulose.[39] The initial plant composition and the interference of various components in the extraction process should be kept in mind, thus, for example, in our previous work, a percentage yield of cellulose of 33.3% was obtained from dead and decaying seagrass, which was attributed to the highly fibrous nature of seagrass, compared to seaweeds, where the major constituent is water.[40]

Fourier transform infrared spectroscopy (FTIR) analysis of cellulose

The FTIR spectra of the celluloses extracted from *Sarconema filliforme* and *Hypnea musciformis* are shown in Figures 3 and 4. The analysis of the spectra revealed major bands between 700 to 3000 cm^{-1}. The samples extracted from *Sarconema filliforme* showed the characteristic bands of cellulose at 619, 1069, 1435, 1646, 2857, 2927 and 3448 cm^{-1}. Among them, the hydrogen bonded -OH stretching vibration was observed at 3448 cm^{-1}, and the weak C-H stretching was observed at 2927 cm^{-1}, respectively. The peak at 1069 cm^{-1} showed the presence of CH_2-O-CH_2 pyranose ring stretching vibration. The asymmetric -CH_2 bending and CH vibration was observed at 1435 cm^{-1}. The cellulose extracted from *Hypnea musciformis* showed the characteristic cellulose bands at 522, 1067, 1432, 1647, 2853, 2925 and 3448 cm^{-1}. Among them, the C-H stretching was observed at 2925 cm^{-1}. The hydrogen bonded -OH stretching was located at 3448 cm^{-1}. The peak at 1067 cm^{-1} was assigned to C-O-C, C-O stretching. The bound H_2O stretching vibration was located at 1647 cm^{-1}. The spectra of the celluloses extracted from both species revealed high intensity peaks at 3448 cm^{-1}, signaling the high number of hydrogen bonds in their structure. These results are in agreement with the findings reported in earlier research.[39,41-44] In addition, non-cellulosic polysaccharides were almost completely eliminated, as indicated by the absence of a peak at 1210 cm^{-1}.

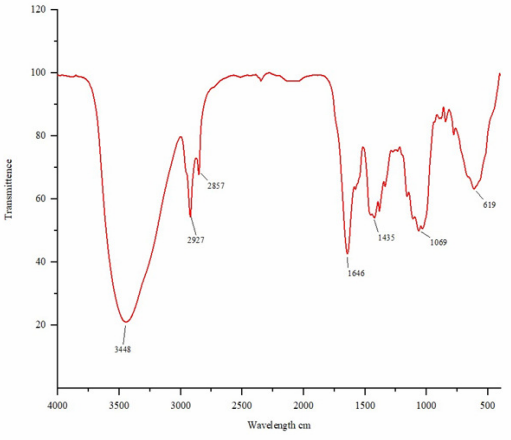

Figure 3: FTIR spectrum of cellulose extracted from *Sarconema filiforme*

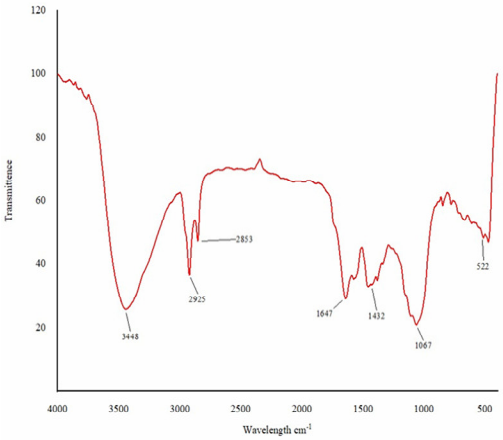

Figure 4: FTIR spectrum of cellulose extracted from *Hypnea musciformis*

X-ray powder diffraction (XRD)

In XRD, the sample is pounded with X rays and the diffraction pattern produced is recorded. The XRD patterns obtained for the two celluloses examined in this study are presented in Figures 5 and 6. In the case of the cellulose extracted from *Sarconema filliforme*, three characteristic diffraction peaks are located around 15.2°, 22.2° and 23.5°, attributed to the crystal planes of cellulose. The major crystalline peak was recorded at 23.5°, confirming the presence of crystalline cellulose. The cellulose of *Hypnea musciformis* showed the characteristic peaks at 2θ = 15.5°, 22.3° and 23.5°, the highest intensity peak being observed at 22.3°. Previous research on rice husk cellulose recorded characteristic diffraction peaks at 2θ = 14.9°, 16.1°, 22.2° and 34.8°, which are quite similar to the present study.[39] No peaks are found in the diffraction patterns of our examined celluloses around 2θ = 19.7°, which is commonly assigned to the less ordered or amorphous region of the cellulose chains.[45,46]

Thermogravimetric analysis (TGA)

The thermogravimetric curves of the celluloses isolated from *Sarconema filliforme* and *Hypnea musciformis* are presented in Figures 7 and 8, respectively, and highlight quite similar thermal behavior of the two celluloses. The thermogravimetric curves disclose three stages of mass loss. The initial mass loss, of 24.1% and 8.2%, for *Sarconema filliforme* and *Hypnea musciformis* celluloses, respectively, occurred up to 150 °C, and can be explained by the evaporation of moisture from the cellulosic

material.[47] The second stage of weight loss was the most significant, with a percentage of mass loss of 32.2% for *Sarconema filliforme* and of 52.2% for *Hypnea musciformis*, and occurred around 250 °C – in this stage, cellulose degradation to charred residue occurs through various processes, such as depolymerization or decomposition of glycosyl units. The final weight loss, of 15.5% for *Sarconema filliforme* and of 23.0% for *Hypnea musciformis*, occurred at temperatures of up to 450 °C and to 500 °C, for the two celluloses, respectively. This final weight loss may be attributed to the breakdown of the charred residue into gaseous products.[48]

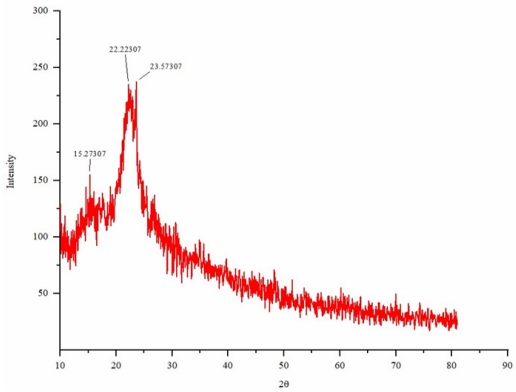

Figure 5: XRD pattern of cellulose extracted from *S. filiforme*

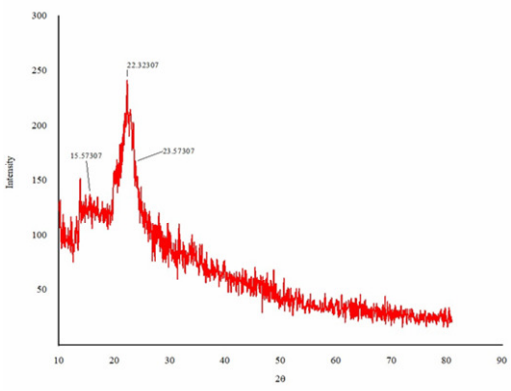

Figure 6: XRD pattern of cellulose extracted from *H. musciformis*

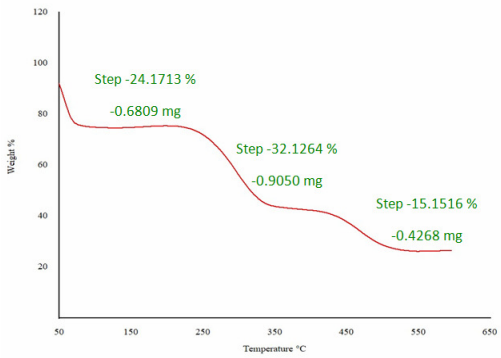

Figure 7: Thermogravimetric curve of cellulose extracted from *S. filiforme*

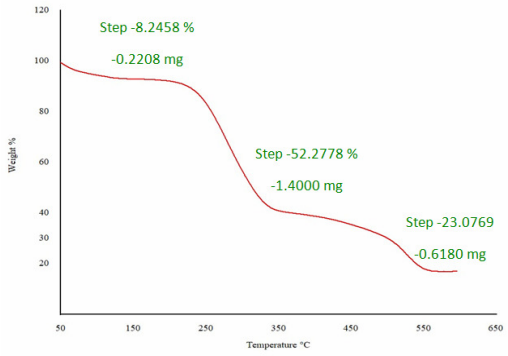

Figure 8: Thermogravimetric curve of cellulose extracted from *H. musciformis*

Scanning electron microscopy (SEM)

The cellulose samples were subjected to SEM to get an insight into their morphology. The micrographs of distinct sections of the celluloses isolated from *Sarconema filliforme* and *Hypnea musciformis*, at various magnifications, are provided in Figures 9 and 10, respectively. As may be observed from the micrographs, the celluloses obtained from both species of seaweeds presented smooth texture, with pore-free, dense morphology. The observation of the smooth surface of the celluloses is consistent with the findings reported in earlier research on other seaweed species. For example, Dalia *et al.*[38] analyzed the surface morphology of cellulose isolated from various seaweed species and noted the smooth surface of numerous filaments assembled into web-like structures. Another study also reported on the smooth texture of cellulose isolated from *Kigelia africana*.[31]

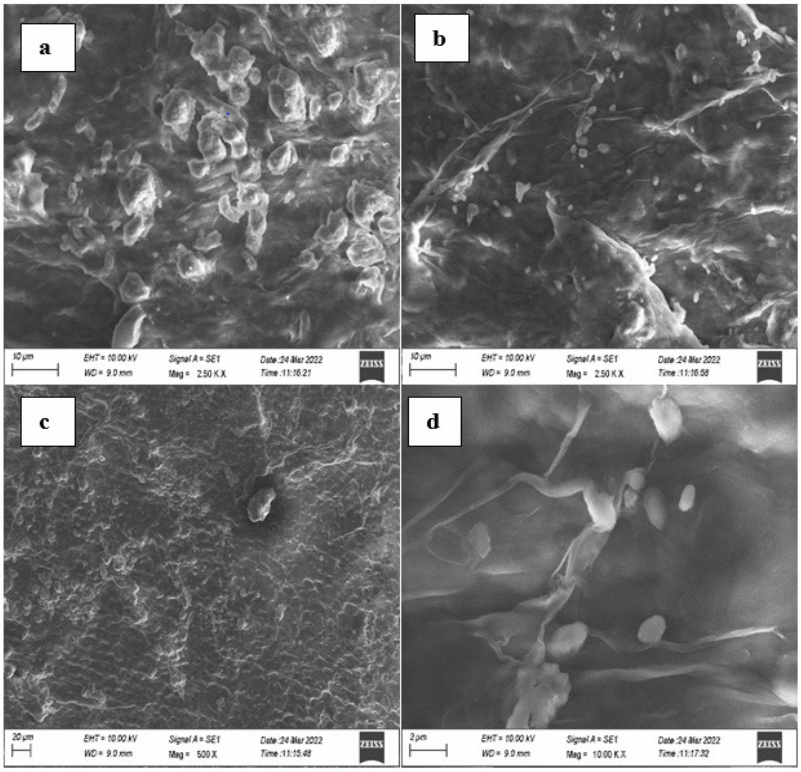

Figure 9: SEM micrographs at different magnifications of cellulose extracted from *S. filiforme*

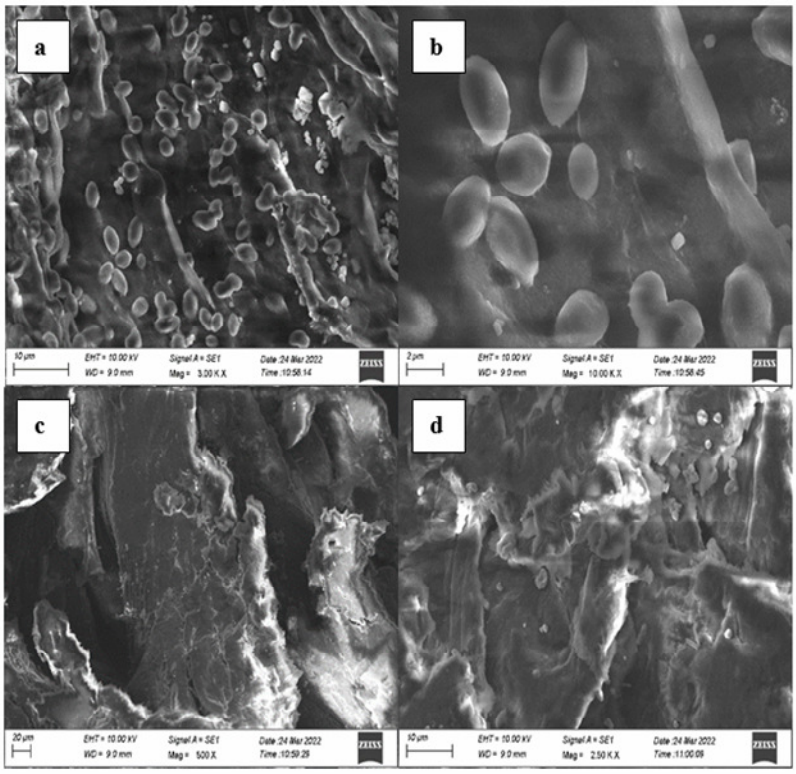

Figure 10: SEM micrographs at different magnifications of cellulose extracted from *H. musciformis*

CONCLUSION

The red seaweed species of *H. musciformis* and *S. filliforme* are commonly found as by-catch on the ropes connecting fishermen's boats to the shore. They are often discarded and regarded as waste. Seaweed samples were used in the present study to extract cellulose in order to turn waste seaweeds into a value-added product. The yield of the cellulose obtained from these samples was very low in this study, but it was in the ranges reported by other authors. The low yields can be considered as normal, being explained by the influence of the chemical composition of these seaweed species. As mentioned earlier, it appears that calcareous Rhodophyta species produce low cellulose amounts, despite their high carbohydrate content, because of their calcium-rich composition. The extracted cellulose was subjected to various characterization studies, using FTIR, XRD, TGA and SEM techniques. FTIR analysis confirmed the presence of characteristic peaks of cellulose in the samples, their crystallinity was confirmed by XRD, while the thermal stability of the cellulose at high temperatures was determined by TGA. The results obtained in this work are in agreement with the findings of previous literature reports. Future research is necessary to reveal the full potential of these seaweed species, and to find out the suitability of the extracted cellulose materials for various applications.

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REFERENCES

[1] M. G. Mehrabani, R. Karimian, B. Mehramouz, M. Rahimi and H. S. Kafil, *Int. J. Biol. Macromol.*, **114**, 961 (2018), http://doi.org/10.1016/j.ijbiomac.2018.03.128

[2] M. G. Mehrabani, R. Karimian, R. Rakhshaei, F. Pakdel, H. Eslami *et al.*, *Int. J. Biol. Macromol.*, **116**, 966 (2018), http://doi.org/10.1016/j.ijbiomac.2018.05.102

[3] B. Chan and K. Leong, *Eur. Spine J.*, **17**, 467 (2008), https://doi.org/10.1007/s00586-008-0745-3

[4] S. Sadreddini, R. Safaralizadeh, B. Baradaran, L. Aghebati-Maleki, M. A. Hosseinpour Feizi *et al.*, *Immunol. Lett.*, **181**, 79 (2018), http://doi.org/10.1016/j.imlet.2016.11.013

[5] A. Sionkowska, *Prog. Polym. Sci.*, **36**, 1254 (2011), http://doi.org/10.1016/j.progpolymsci.2011.05.003

[6] T. Ozawa, D. A. Mickle, R. D. Weisel, N. Koyama, S. Ozawa *et al.*, *Circulation*, **106**, 176 (2002), https://doi.org/10.1161/01.cir.0000032901.55215.cc

[7] L. M. Cross, A. Thakur, N. A. Jalili, M. Detamore and A. K. Gaharwar, *Acta Biomater.*, **42**, 2 (2016), http://doi.org/10.1016/j.actbio.2016.06.023

[8] M. S. Zafar, A. A. Alnazzawi, M. Alrahabi, M. A. Fareed, S. Najeeb *et al.*, *Int. J. Mol. Sci.*, **20**, 5960 (2019), http://doi.org/10.3390/ijms20235960

[9] S. Raveendran, A. K. Rochani, T. Maekawa and D. S. Kumar, *Materials*, **10**, 929 (2013), http://doi.org/10.3390/ma10080929

[10] S. Gustafson, *Glycobiology*, **7**, 1209 (1997), http://doi.org/10.1093/glycob/7.8.1209

[11] C. R. Parish, C. Freeman, K. J. Brown, D. J. Francis and W. B. Cowden, *Cancer Res.*, **15**, 3433 (1999)

[12] L. S. Costa, G. P. Fidelis, S. L. Cordeiro, R. M. Oliveira, D. A. Sabry *et al.*, *Int. J. Mol. Sci.*, **12**, 3352 (2011), http://doi.org/10.3390/ijms12053352

[13] R. Jayakumar, D. M. Manzoor, S. V. Nair and H. Tamura, *Carbohyd. Polym.*, **82**, 227 (2010), http://doi.org/10.1016/j.carbpol.2010.04.074

[14] T. Heinze and T. Liebert, *Prog. Polym. Sci.*, **26**, 16 (2001), http://doi.org/10.1016/S0079-6700(01)00022-3

[15] D. Klemm, B. Heublein, H. P. E. Habil and A. Bohan, *J. German Chem. Soc.*, **44**, 3358 (2005), http://dx.doi.org/10.1002/anie.200460587

[16] M. Q. Zhang, *Express Polym. Lett.*, **1**, 406 (2007), http://doi.org/10.3144/expresspolymlett.2007.57

[17] L. Yan, W. Li, Z. Qi and S. Liu, *J. Polym. Res.*, **13**, 375 (2006), https://doi.org/10.1007/s10965-006-9054-x

[18] A. Biswas, R. L. Shogren and J. L. Willett, *Biomacromolecules*, **6**, 1843 (2005), https://doi.org/10.1021/bm0501757

[19] D. G. Yu, J. H. Yu, L. Chen, G. R. Williams and X. Wang, *Carbohyd. Polym.*, **90**, 1016 (2012), http://dx.doi.org/10.1016/j.carbpol.2012.06.036

[20] D. G. Yu, X. Y. Li, X. Wang, W. Chian, Y. Z. Liao *et al.*, *Cellulose*, **20**, 379 (2013), https://doi.org/10.1007/s10570-012-9824-z

[21] M. Ilangovan, V. K. Guna, H. Chunyan, G. S. Nagananda and N. Reddy, *Ind. Crop. Prod.*, **112**, 556 (2018), https://doi.org/10.1016/j.indcrop.2017.12.042

[22] S. G. Jebadurai, R. E. Raj, V. S. Sreenivasan and J. S. Binoj, *Carbohyd. Polym.*, **207**, 675 (2019), https://doi.org/10.1016/j.carbpol.2018.12.027

[23] M. Kathirselvam, A. Kumaravel, V. P. Arthanarieswaran and S. S. Saravanakumar, *Carbohyd.*

Polym., **212**, 439 (2019), https://doi.org/10.1016/j.carbpol.2019.02.072
[24] N. Reddy and Y. Yang, *Bioresour. Technol*., **100**, 3563 (2009), https://doi.org/10.1016/j.biortech.2009.02.047
[25] C. S. R. Freire, S. C. M. Fernandes, A. J. D. Silvestre and C. Pascoal Neto, *Holzforschung*, **67**, 603 (2013), https://doi.org/10.1515/hf-2012-0127
[26] J. Shojaeiarani, D. S. Bajwa and S. Chanda, *Compos. Part C*, **5**, 100164 (2021), https://doi.org/10.1016/j.jcomc.2021.100164
[27] Q. He, Q. Wang, H. Zhou, R. Dandan, H. Yunhai *et al*., *Cellulose*, **25**, 5523 (2018), https://doi.org/10.1007/s10570-018-1966-1
[28] K. Santhanam, A. Kumaravel, S. S. Saravanakumar and V. P. Arthanarieswaran, *Int. J. Polym. Anal. Charact*., **21**, 267 (2016), https://doi.org/10.1080/1023666X.2016.1147654
[29] A. Trivedi, P. S. Mavi, D. Bhatt and A. Kumar, *Nat. Commun*., **7**, 11392 (2016), https://doi.org/10.1038/ncomms11392
[30] I. Manikandan, G. Vijaykumar, B. Prajwal, J. Qiuran and R. Narendra, *Carbohyd. Polym.*, **236**, 115996 (2020), https://doi.org/10.1016/j.carbpol.2020.115996
[31] S. Y. Nair, F. Nauer and C. O. Mariana, *J. Appl. Phycol*., **32**, 3585 (2020), https://doi.org/10.1007/s10811-020-02209-x
[32] M. Szymanska-Chargot, M. Chylinska, K. Gdula, A. Koziol and A. Zdunek, *Polymers (Basel)*, **9**, 495 (2017), https://doi.org/10.3390%2Fpolym9100495
[33] A. K. Siddhantaa, K. Prasad, R. Meena, P. Gayatri, G. K. Mehta *et al*., *Bioresour. Technol*., **100**, 6669 (2009), https://doi.org/10.1016/j.biortech.2009.07.047
[34] N. Wahlström, U. Edlund, H. Pavia, T. Gunilla, J. Alaksander *et al*., *Cellulose*, **27**, 3707 (2020), https://doi.org/10.1007/s10570-020-03029-5
[35] A. K. Siddhanta, K. Sanjay, G. K. Mehta, M. U. Chhatbara, M. D. Oza *et al*., *Nat. Prod. Commun*., **8**, 497 (2013), https://doi.org/10.1177/1934578X1300800423
[36] A. K. Siddhanta, K. Sanjay, G. K. Mehta, M. U. Chhatbara, M. D. Oza *et al*., *J. Appl. Phycol*., **23**, 919 (2011), https://doi.org/10.1007/s10811-010-9599-2
[37] G. F. El-Said and A. El-Sikaily, *Environ. Monit. Assess*., **7**, 6089 (2013), https://doi.org/10.1007/s10661-012-3009-y
[38] M. S. A. S. Dalia and M. Mona Ismail, *Egypt. J. Aqua. Res*., (in press) (2021), https://doi.org/10.1016/j.ejar.2021.11.001
[39] M. D. Archana, A. A. Abdul and M. P. Hazarika, *Carbohyd. Polym*., **112**, 342 (2014), https://doi.org/10.1016/j.carbpol.2014.06.006
[40] R. Varma and V. Sugumar, *Cellulose Chem. Technol*., **56**, 39 (2022), https://doi.org/10.35812/CelluloseChemTechnol.2022.56.03
[41] J. X. Sun, F. Xu, X. F. Sun, B. Xiao and R. C. Sun, *Polym. Degrad. Stab*., **88**, 521 (2005), https://doi.org/10.1016/j.polymdegradstab.2004.12.013
[42] D. S. Lakshmi, N. Trivedi and C. R. K. Reddy, *Carbohyd. Polym*., **157**, 1604 (2017), https://doi.org/10.1016/j.carbpol.2016.11.042
[43] C. S. R. Freire, A. J. D. Silvestre, C. P. Neto, M. N. Belgacem and A. Gandini, *J. Appl. Polym. Sci*., **100**, 1093 (2006), https://doi.org/10.1002/app.23454
[44] W. Hu, S. Chen, Q. Xu and H. Wang, *Carbohyd. Polym*., **83**, 1575 (2011), https://doi.org/10.1016/j.carbpol.2010.10.016
[45] B. Deepa, E. Abraham, N. Cordeiro, M. Mozetic, A. P. Mathew *et al*., *Cellulose*, **22**, 1075 (2015), https://doi.org/10.3390/ma9010050
[46] M. Roman and W. T. Winter, *Biomacromolecules*, **5**, 1671 (2004), https://doi.org/10.1021/bm034519+
[47] N. Wang, E. Ding and R. Cheng, *Polymer*, **48**, 3486 (2007), https://doi.org/10.1016/j.polymer.2007.03.062
[48] D. R. Mulinari, H. J. C. Voorwald, M. O. H. Cioffi, M. L. C. P. da Silva, T. G. da Cruz *et al*., *Compos. Sci. Technol*., **69**, 214 (2009), https://doi.org/10.1016/j.compscitech.2008.10.006