Papermaking properties of bacterial nanocellulose produced from mother of vinegar, a waste product after classical vinegar production

GREGOR LAVRIČ, DAŠA MEDVEŠČEK, AND MATEJ SKOČAJ

ABSTRACT: Bacterial nanocellulose (BNC) has gained a lot of attention in recent years due to its nano-size-derived properties. Although it is essentially chemically similar to plant-derived cellulose, it has smaller size and is enriched in free hydroxyl groups, which greatly improve mechanical properties of reinforced paper. However, although BNC has some unique features, it comes at a high price. In this paper, we introduce a new solution for BNC production. We have isolated bacterial nanocellulose directly from agro-industrial waste—mother of vinegar—and used it in the production of paper sheets. We show here that paper sheets made with the addition of only 10% bacterial nanocellulose from mother of vinegar substantially improved basic mechanical as well as printing properties of paper.

Application: In this paper, we have demonstrated the simple isolation process of BNC, as well as summarized how the basic paper properties of paper sheets are improved with addition of BNC.

Nanocellulose is a term that it is used for cellulose of nanoscale dimensions. Nanocellulose includes nanofibers, and nanocrystals, and it is routinely produced from plant-derived cellulose by a variety of chemical and mechanical procedures. Bacterial nanocellulose (BNC) represents a special type of nanocellulose that is produced by various bacterial genera [1-5]. Among these, the Gram-negative bacterium *Acetobacter xylinum*, which typically lives on rotten fruit, was described as the most effective producer of BNC [2].

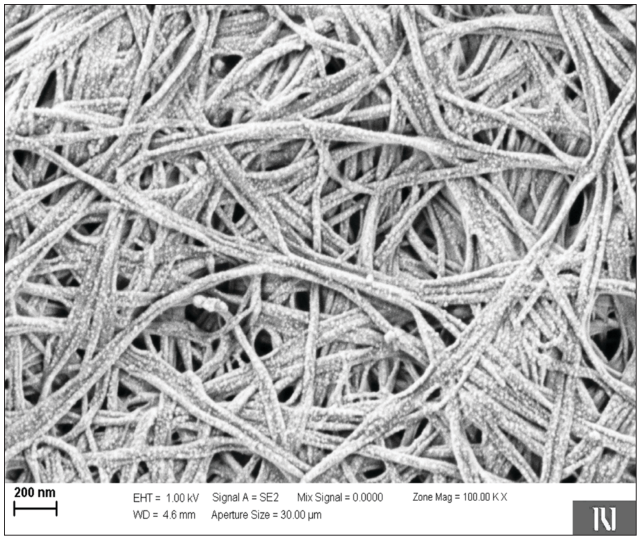
Bacterial nanocellulose can be industrially produced by static or shaking culture methods. However, as the production costs are relatively high, and the yields of BNC are relatively low, large-scale production of BNC has not been feasible to date [6,7]. Therefore, a lot of effort has been put into studies of the solutions for large-scale BNC production. New forms of bioreactors have been explored, with different strains of bacteria tested. Some of these bacterial strains have undergone genetic manipulation in an effort to increase BNC yields [8-11]. Furthermore, as the medium for growing the bacteria is the most important factor in terms of the costs of BNC production, studies have focused on the use of alternative carbon sources and feedstock, as well as different supplements [12-19]. BNC is chemically similar to plant cellulose, but it has a smaller fiber size and does not contain lignin, hemicellulose, or pectin; therefore, it represents an exceptionally pure source of cellulose [7,20-26]. Furthermore, as well as having a uniform nature, BNC has a high degree of crystallinity, a large specific surface area, and reinforcing potential, and also high density, elasticity, water-absorbing capacity, and specific stiffness and strength [27]. If compared to plant-derived cellulose, BNC is in a nano-form, and as it is very pure, it does not require delignification or demanding chemical or mechanical procedures for its extraction.

Due to its exceptional mechanical properties, BNC is already used in several industries (e.g., biomedicine, construction, textile, waste treatment, and electrical products) and has many interesting proposed applications [28,29]. In recent years, literature reports have demonstrated its use in the papermaking industry. As the global demand for protecting forests has increased, the recycling of paper and the exploitation of non-woody fibers are possible solutions towards the protection of the world's forests [30]. However, recycled fibers have reduced mechanical and physical properties compared to primary fibers, which is a direct consequence of the recycling procedures [31-34].

The small size and enrichment in free hydroxyl groups in BNC can improve the characteristics of paper made from recycled or lower-grade quality fiber [27]. The use of BNC in papermaking can also result in improved durability of pulp when integrated into paper, as well as protecting and improving the surface of the paper [35-39]. Therefore, BNC has a promising role in papermaking, which allows for the creation of flexible and durable paper, as well as cardboard and paper with high filler content [7,36,40-42]. BNC is also a promising material for the packing sector, as it can increase the barrier properties of paper (e.g., impermeability to microorganisms, fats, oils, and oxygen), and can therefore be used as a supplement for plastics in the packing sector [43-45].

Some recent studies have demonstrated that BNC can be used for restoration of damaged paper and as a paper-reinforcing agent, as it can improve the properties of paper made from non-woody and low-grade fiber sources. Furthermore, it can be used to improve paper quality by direct application to the paper surface, and it can even improve the fire resistance of paper [30,40,42-49]. To conclude, BNC enrichment of paper improves the quality of the paper in terms of higher density, stiffness, durability, tear index, burst index, tensile index, permeability, optical properties, and mechanical strength [27].

The properties of BNC-enriched paper have been extensively documented, but to date, no BNC-enriched paper has been made using mother of vinegar, a low-cost agro-industrial waste that is produced during traditional vinegar making. The mother of vinegar is a biofilm produced during traditional vinegar production. It is chemically identical to laboratory-produced BNC. The main difference between both types of the biofilm is that that traditional vinegar fermentation is not initiated through the inoculation of acetic bacteria as starter cultures. Within this short note, we demonstrate the properties of paper enriched with BNC derived from the biofilm that is a byproduct from traditional vinegar fermentation.



1. Scanning electron microscopy image of bacterial nanocellulose (BNC), as derived from the biofilm after acetic fermentation of apple juice.

MATERIALS AND METHODS

The source of BNC in this study was the cellulose-rich biofilm that was formed after acetic fermentation of apple juice and was obtained from a local vinegar producer. After 80 days of spontaneous acetic fermentation in a covered 50-L container, the biofilm was formed. The mother of vinegar itself contained some impurities because pulped apples were used for the acetic fermentation. Therefore, the main impurities were brownish-colored particles of the apple pulp that had served as the raw material for the vinegar production. To remove these impurities, the biofilm was initially washed under running water for 2 min. To aid in further handling, the biofilm was cut into round "mats' of ~10-cm diameter and ~1.5-cm thickness. These biofilm mats had a dry content of 15% before further processing.

The biofilm mats were thermally treated (60°C) in 50% (v/v) aqueous (distilled water) ethanol (Pharmachem Laboratories; Kearny, NJ, USA) for 40 min in a 2-L beaker under magnetic stirring. This was followed by thermochemical treatment with 0.1 M sodium hydroxide (NaOH, Merck; Kenilworth, NJ, USA) under the same conditions. The biofilm mats were processed in sets of four.

The biofilm mats were then disintegrated at 7000 rpm for 1 min using a Kinematica Polytron PT45/80 (Kinematica AG; Luzern, Switzerland) to allow removal of the impurities that were held within the structure of the biofilm mats. During the disintegration, the homogeneous structure of the biofilm mats was completely broken up. Next, the disintegrated biofilm suspension was treated with 0.1 M NaOH (40 min, at 60°C, under magnetic stirring) and washed with 1.5 L of distilled water, followed by further homogenization (7000 rpm, 6 min) in distilled water. This separated the individual nanofibrils (**Fig. 1**), resulting in the formation of a homogeneous transparent BNC gel.

To determine the influence of the BNC obtained on the properties of paper, paper sheets were made according to the international standard ISO 5269-2:2004 "Pulps—Preparation of laboratory sheets for physical testing—Part 2: Rapid-Köthen method," using a Frank-PTI (Birkenau, Germany) sheet former. Paper sheets were produced with eucalyptus pulp (Ence Navia; Navia, Spain) and 0.5% cationic starch (Papiran SKM-42, Helios Kemostik; Kamnik, Slovenia), which served as the retention agent. Paper sheets without BNC were made as the reference sample (Reference). Further sheets of paper were made that contained 10% (BNC 10) and 20% (BNC 20) (w/w) BNC, according to the weight of the dry matter of the basic fibers. Before testing, all of the paper samples were conditioned according to ISO 187:1990 "Paper, board and pulps—Standard atmosphere for conditioning and testing and procedure for monitoring the atmosphere and conditioning of samples." The tests were carried out under 50% relative humidity at 23°C. The following properties of the final paper sheets were determined according to standard paper test methods described in **Table I**: grammage, density, opacity, thickness, air permeance, roughness, tear index, burst index, tensile index, and water penetration. Laboratory paper sheets were also printed with water-based printing ink, using a flexography printability tester IGT F1 (IGT Testing Systems;

Property	Test Method	Test Name
Grammage	ISO 536:2012	Paper and board—Determination of grammage
Density	ISO 534:2011	Paper and board—Determination of thickness, density and specific volume
Opacity	ISO 2471:2008	Paper and board—Determination of opacity (paper backing)—Diffuse reflectance method
Thickness	ISO 534:2011	Paper and board—Determination of thickness, density and specific volume
Air permeance	ISO 5636-3:2013	Paper and board—Determination of air permeance (medium range)—Part 3: Bendtsen method
Roughness	ISO 8791-2:2013	Paper and board—Determination of roughness/smoothness (air leak methods)—Part 2: Bendtsen method
Tear index	ISO 1974:2012	Paper—Determination of tearing resistance—Elmendorf method
Burst index	ISO 2758:2014	Paper—Determination of bursting strength
Tensile index	ISO 1924-2:2008	Paper and board—Determination of tensile properties—Part 2: Constant rate of elongation method (20 mm/min)
Water penetration	emtec PDA.C 02 device (emtec Electronic; Leipzig, Germany)	

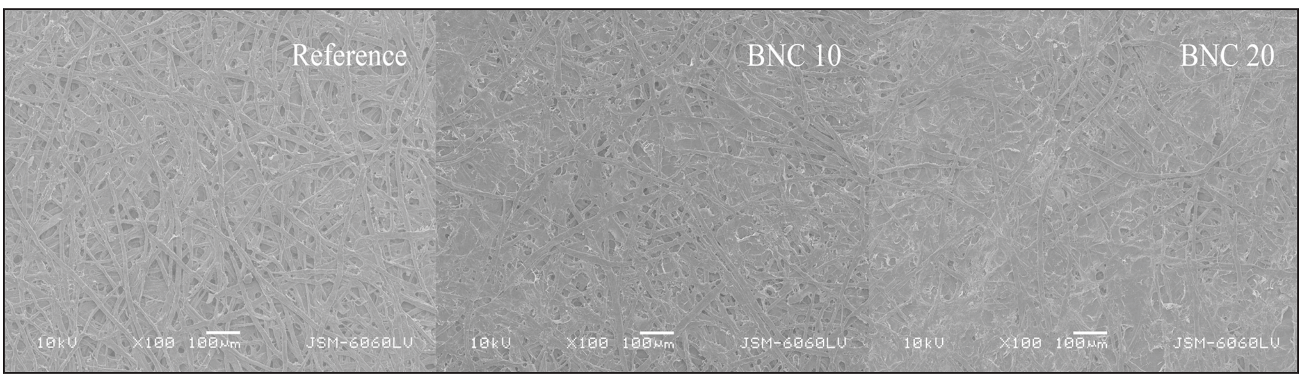
I. Test methods used to determine properties of the final paper sheets.

Almere, Netherlands). An anilox printing cylinder with lineature of 180 lines per centimetre and transfer volume of 4.8 g m^{-2} was used for printing. Printing speed was 0.6 m s^{-1}, the pressure of the anilox cylinder to printing plate was 60 N, and the printing pressure was set at 90 N.

RESULTS AND DISCUSSION

The bacterial nanocellulose filled the porous structure of the paper (**Fig. 2**), and thereby increased the grammage, density, and opacity of the paper, although it did not significantly affect the thickness of the paper samples (**Table II**). The increase in grammage and density of the BNC-enriched paper sheets were not surprising and were also already described in related studies [43,48,49]. For the same reason, the air permeability of the paper significantly decreased (Table II).

Furthermore, BNC enrichment of the paper decreased the roughness of the paper, which in turn positively affected the printing properties of the paper. The results of print sharpness, determined by Modulation Transfer Function, which is in accordance with ISO 12233:2017 "Photography—Electronic still picture imaging—Resolution and spatial frequency responses," show substantial improvement (Table II). The BNC-enriched paper had reduced po-



2. Scanning electron microscope images of paper surfaces, with addition of 10% BNC (BNC 10, middle); 20% BNC (BNC 20, right), and without BNC (Reference, left).

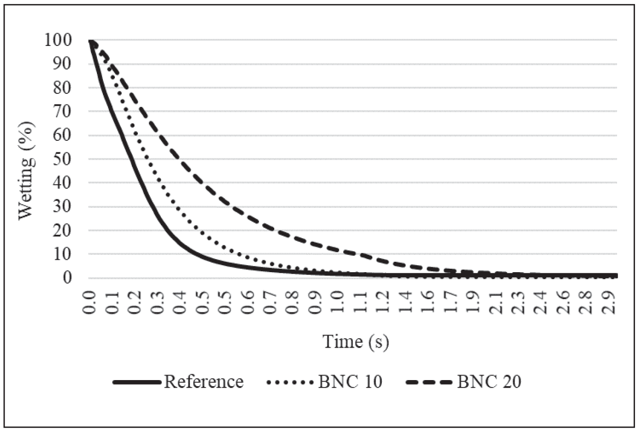
	Reference Sample	BNC 10	BNC 20
Grammage, g m^{-2}	66.8 ± 0.341	71.3 ± 0.673	77.0 ± 0.739
Density, kg m^{-3}	631 ± 6.141	666 ± 10.478	713 ± 8.869
Opacity	73.36 ± 0.925	75.07 ± 0.525	75.66 ± 0.435
Thickness, mm	0.106 ± 0.002	0.107 ± 0.001	0.108 ± 0.001
Air permeance	2250 ± 138	240 ± 20	40 ± 7
Roughness, mL min^{-1}	480 ± 27	380 ± 15	360 ± 22
Print sharpness, mm l^{-1}	99.18 ± 4.759	104.77 ± 6.030	109.06 ± 4.545
Optical density values	0.635 ± 0.020	0.710 ± 0.015	0.721 ± 0.021
Tear index	7.5 ± 0.104	7.9 ± 0.154	8.2 ± 0.160

II. Grammage, density, opacity, thickness, air permeance, roughness, print sharpness, optical print density values and tear index of reference and bacterial nanocellulose (BNC)-enriched samples.

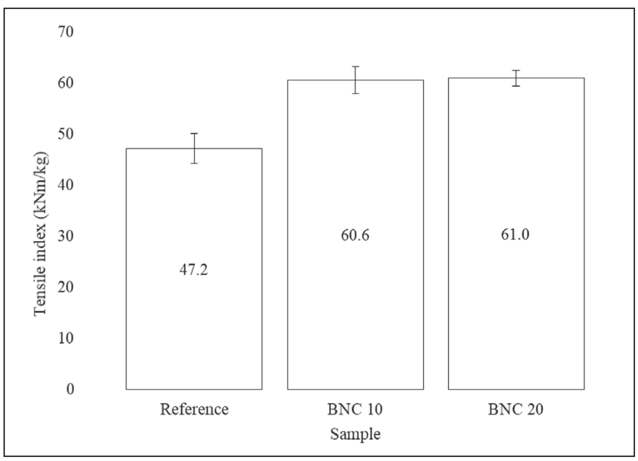
rosity and lower paper roughness, leading to better retention of the pigment particles of the printing ink on the paper surface, which increased the optical density values of the prints, measured in accordance with ISO 5-4:2009 "Photography and graphic technology—Density measurements—Part 4: Geometric conditions for reflection density" (Table II). It is known that paper roughness can result in micro spots of the reflected light that are integrated with the ink color by the eye. This results in "dilution" of the color, which, in turn, results in reduced ink density and print contrast. Smooth, uniform paper is especially important for high-quality image reproduction [50]. Due to its higher density, the paper enriched with BNC provides better barrier properties, and because it has a smoother surface, it is also more suitable for printing. The improvements of both of these properties are especially important for packaging paper. The increased sheet density is also a consequence of increased amount of links between paper fibers, which leads to slightly higher values of the tear index, but has no drastic effect on the burst index of the samples (data not shown).

Figure 3 shows the dynamic penetration of water into the paper structure as a function of time. The reference sample achieved faster penetration of water compared to BNC-enriched samples, which is reflected in the slower curves of the BNC 10 and BNC 20. These results go well in hand with the higher density and lower porosity values of the BNC-enriched paper samples.

Bacterial nanocellulose had a positive effect on the tensile index of the paper (**Fig. 4**). The enrichment of the paper with 10% and 20% of BNC increased the tensile indices by ~30%. However, as there was no significant increase in the tensile index of the paper when the BNC addition was increased from 10% to 20%, we can conclude that 10% BNC was enough to fill all the available pores in the paper structure. A higher tensile index is a sign that



3. Dynamic penetration of water into the paper structure as a function of time with no BNC, 10% BNC, and 20% BNC.



4. Influence of BNC on the tensile index of the paper sheets produced. Comparison of the paper made from eucalyptus pulp without BNC (Reference) and with addition of 10% BNC and 20% BNC. Data are represented as means ± standard deviation from two individual experiments. Each experiment included 10 samples (n = 20 per sample).

BNC-enriched paper would have better physical strength at a similar grammage or mass. The increase in the physical strength of the BNC-enriched papers is mainly due to the larger number of new hydrogen bonds established in the structure of the paper. A high tensile index is particularly important for packaging papers, which need to have the best physical properties possible at the lowest possible grammage or mass.

CONCLUSIONS

The outstanding and unique properties of BNC have now been known for some time. The particular properties of this natural and biodegradable nanomaterial have opened the door for its use in many different industries. The use of BNC for papermaking is of great importance, as it influences the paper quality in many different ways. Although some studies have investigated the influence of laboratory-produced BNC on paper-based products, to the best of our knowledge, this is the first report where mother of vinegar, a byproduct after classical vinegar fermentation, has been used as the source of BNC.

In this paper, we have shown that BNC can be produced from mother of vinegar, which is a waste product after production of vinegar, and that it has positive effects on the basic mechanical and printing properties of paper sheets. This agro-industrial waste might not be appropriate for applications in the medical field, but we have shown here that it behaves essentially identically to industrially-produced nanocellulose. Furthermore, as nanocellulose and BNC are of great importance for different industries, their price has constantly increased. This BNC produced from mother of vinegars could therefore represent a cheaper substitute for this costly material. Finally, this reuse of various resources meets the goals for a circular economy where nothing is wasted, and at the same time, it helps to protect our environment.

To conclude, we have shown here that: (i) BNC isolated from mother of vinegar has the potential for use in the papermaking industry, since (ii) it improves basic mechanical characteristics; and (iii) improves printing properties of paper sheets. Although laboratory-made BNC has been used in many studies, to the best of our knowledge, this is the first report where the mother of vinegar, a byproduct after classical vinegar fermentation, has been used as the source of BNC to improve the printing properties of paper sheets. **TJ**

ACKNOWLEDGEMENTS

The study was carried out within the RDI project Cel.Krog: "Potential of biomass for development of advanced materials and bio-based products" (contract number: OP20.00365), and was co-financed by the Republic of Slovenia, Ministry of Education, Science and Sport, and the European Union, under the European Regional Development Fund, 2016–2020. We also thank the Slovenian Research Agency for financial support (Grant P1-0207).

LITERATURE CITED

1. Ross, P., Mayer, R., Benziman, M., *Microbiol. Rev.* 55: 35(1991). https://doi.org/10.1128/MMBR.55.1.35-58.1991.
2. Shoda, M. and Sugano, Y., *Biotechnol. Bioprocess Eng.* 10: 1(2005). https://doi.org/10.1007/BF02931175.
3. Jung, J.Y., Khan, T., Park, J.K., et al., *Korean J. Chem. Eng.* 24: 265(2007). https://doi.org/10.1007/s11814-007-5058-4.
4. Yamada, Y., Yukphan, P., Lan Vu, H.T., et al., *J. Gen. Appl. Microbiol.* 58: 397(2012). https://doi.org/10.2323/jgam.58.397.
5. Lee. K-Y., Buldum, G., Mantalaris, A., et al., *Macromol. Biosci.* 14: 10(2014). https://doi.org/10.1002/mabi.201300298.
6. Chao, Y., Ishida, T., Sugano, Y., et al., *Biotechnol. Bioeng.* 68: 345(2000). https://doi.org/10.1002/(SICI)1097-0290(20000505)68:3<345::AID-BIT13>3.0.CO;2-M.
7. Chawla, P.R., Bajaj, I.B., Survase, S.A., et al., *Food Technol. Biotechnol.* 47: 107(2009).
8. Cheng, H-P., Wang, P-M., Chen, J-W., et al., *Biotechnol. Appl. Biochem.* 35: 125(2002). https://doi.org/10.1042/BA20010066.
9. Song, H-J., Li, H., Seo, J-H., et al., *Korean J. Chem. Eng.* 26: 141(2009). https://doi.org/10.1007/s11814-009-0022-0.
10. Kralisch, D., Hessler, N., Klemm, D., et al., *Biotechnol. Bioeng.* 105: 740(2010).
11. Rajwade, J.M., Paknikar, K.M., and Kumbhar, J.V., *Appl. Microbiol. Biotechnol.* 99: 2491(2015). https://doi.org/10.1007/s00253-015-6426-3.
12. Masaoka, S., Ohe, T., and Sakota, N., *J. Ferment. Bioeng.* 75: 18(1993). https://doi.org/10.1016/0922-338X(93)90171-4.
13. Matsuoka, M., Tsuchida, T., Matsushita, K., et al., *Biosci., Biotechnol., Biochem.* 60: 575(1996). https://doi.org/10.1271/bbb.60.575.
14. Yang, Y.K., Park, S.H., Hwang, J.W., *J. Ferment. Bioeng.* 85: 312(1998). https://doi.org/10.1016/S0922-338X(97)85681-4.
15. Ishihara, M., Matsunaga, M., Hayashi, N., et al., *Enzyme Microb. Technol.* 31: 986(2003). https://doi.org/10.1016/S0141-0229(02)00215-6.
16. Son, H-J., Kim, H-G., Kim, K-K., et al., *Bioresour. Technol.* 86: 215(2003).
17. Keshk, S.M.A.S. and Sameshima, K., *Afr. J. Biotechnol.* 4: 478(2005).
18. Zhou, L.L., Sun, D.P., and Hu, L.Y., *J. Ind. Microbiol. Biotechnol.* 34: 483(2007). https://doi.org/10.1007/s10295-007-0218-4.
19. Mikkelsen, D., Flanagan, B.M., Dykes, G.A., et al., *J. Appl. Microbiol.* 107: 576(2009). https://doi.org/10.1111/j.1365-2672.2009.04226.x.
20. Yoshinaga, F., Tonouchi, N., and Watanabe, K., *Biosci., Biotechnol., Biochem.* 61: 219(1997). https://doi.org/10.1271/bbb.61.219.
21. Jonas, R. and Farah, L.F., *Polym. Degrad. Stab.* 59: 101(1998). https://doi.org/10.1016/S0141-3910(97)00197-3.
22. Schrecker, S.T. and Gostomski, P.A., *Biotechnol. Lett.* 27: 1435(2005). https://doi.org/10.1007/s10529-005-1465-y.
23. El-Saied, H., El-Diwany, A.I., Basta, A.H., et al., *BioResources* 3: 1196(2008).

24. Fang, L. and Catchmark, J.M., *Cellulose* 21: 3965(20014). https://doi.org/10.1007/s10570-014-0443-8.

25. Jeon, S., Yoo, Y-M., Park, J-W., et al., *Curr. Appl. Phys.* 14: 1621(2014). https://doi.org/10.1016/j.cap.2014.07.010.

26. Rosa, J.R., da Silva, I.S.V., de Lima, C.S.M., et al., *Cellulose* 21: 1361(2014). https://doi.org/10.1007/s10570-014-0169-7.

27. Skočaj, M., *Cellulose* 26: 6477(2019). https://doi.org/10.1007/s10570-019-02566-y.

28. Lu, Y., Tekinlap, H., Eberle, C.C., et al., *TAPPI J.* 13(6): 47(2014). https://doi.org/10.32964/TJ13.6.47.

29. Lindstrom, T., *TAPPI J.* 18(5): 308(2019). https://doi.org/10.32964/TJ18.5.308.

30. Xiang, Z., Jin, X., Liu, Q., et al., *Cellulose* 24: 5147(2017a). https://doi.org/10.1007/s10570-017-1468-6.

31. Blanco, A., Miranda, R., and Monte, M.C., *For. Syst.* 22: 471(2013). https://doi.org/10.5424/fs/2013223-03677.

32. Kose, R., Yamaguchi, K., and Okayama, T., *Cellulose* 23: 1393(2016). https://doi.org/10.1007/s10570-016-0874-5.

33. Laftah, W.A. and Rahman, W.A.W.A., *J. Nat. Fibers* 13: 85(2016). https://doi.org/10.1080/15440478.2014.984060.

34. Campano, C., Merayom, N., Balea, A., et al., *Cellulose* 25: 269(2018a). https://doi.org/10.1007/s10570-017-1552-y.

35. Goncalves, M. and Łaszkiewicz, B., *Prz. Pap. R.* 55: 657(1999).

36. Iguchi, M., Yamanaka, S., Budhiono, A., *J. Mater. Sci.* 35: 261(2000). https://doi.org/10.1023/A:1004775229149.

37. Serafica, G., Mormino, R., and Bungay, H., *Appl. Microbiol. Biotechnol.* 58: 756(2002). https://doi.org/10.1007/s00253-002-0978-8.

38. Mormino, R. and Bungay, H., *Appl. Microbiol. Biotechnol.* 62: 503(2003). https://doi.org/10.1007/s00253-003-1377-5.

ABOUT THIS PAPER

Cite this article as:
Lavrič, G., Medveščeck, D., and Skočaj, M., *TAPPI J.* 19(4): 197(2020). https://doi.org/10.32964/TJ19.4.197.

DOI: https://doi.org/10.32964/TJ19.4.197

ISSN: 0734-1415

Publisher: TAPPI Press

About this journal

39. Presler, S. and Surma-Ślusarska, B., *Przem. Chem.* T85(8-9): 1297(2006).

40. Basta, A.H. and El-Saied, H., *J. Appl. Microbiol.* 107: 2098(2009). https://doi.org/10.1111/j.1365-2672.2009.04467.x.

41. Ashjaran, A., Yazdanshenas, M.E., Rashidi, A., et al, *J. Text. Inst.* 104: 121(2013). https://doi.org/10.1080/00405000.2012.703796.

42. Fillat, A., Martínez, J., Valls, C., et al., *Cellulose* 25: 6093(2018). https://doi.org/10.1007/s10570-018-1967-0.

43. Gao, W-H., Chen, K-F., Yang, R-D., et al., *BioResources* 6: 144(2011).

44. Santos, S.M., Carbajo, J.M., and Go'mez, N., et al., *J. Mater. Sci.* 52: 5882(2017). https://doi.org/10.1007/s10853-017-0824-0.

45. Xiang, Z., Liu, Q., Chen, Y., et al., *Cellulose* 24: 3513(2017b). https://doi.org/10.1007/s10570-017-1361-3.

46. Santos, S.M., Carbajo, J.M., Quintana, E., et al., *Carbohydr. Polym.* 116: 173(2015). https://doi.org/10.1016/j.carbpol.2014.03.064.

ABOUT THE AUTHORS

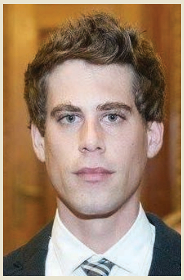
This topic was chosen for research because there is much waste of valuable and diverse molecules that could contribute to green growth and a circular economy where nothing is wasted. Through our research, we were surprised to find how few in the research community and industry have embraced this concept.

Upon review of the literature, we found that no one had considered mother of vinegar as a source for nanocellulose, yet our research showed that this waste product is a valid source of bacterial nanocellulose (BNC). Mother of vinegar could serve as a cheap and abundant source of nanocellulose, which is becoming an important substance in papermaking.

Our next step is to compare the behavior of commercial nanocellulose, such as nanocrystalline cellulose (NCC) and nanofibrillated cellulose (NFC), with the behavior of our isolated BNC from mother of vinegar in different paper-based applications.

Lavrič

Medveščcek



Skočaj

Lavrič is young researcher and Medveščcek is researcher at the Pulp and Paper Institute in Ljubljana, Slovenia. Skočaj is teaching assistant, Biotechnical Faculty, in the Department of Biology at the University of Ljubljana, in Ljubljana, Slovenia. Email Skočaj at matej.skocaj@bf.uni-lj.si.

47. Santos, S.M., Carbajo, J.M., Go’mez, N., et al., *J. Mater. Sci.* 51: 1553(2016). https://doi.org/10.1007/s10853-015-9477-z.

48. Tabarsa, T., Sheykhnazari, S., and Ashori, A., *Int. J. Biol. Macromol.* 101: 334(2017). https://doi.org/10.1016/j.ijbiomac.2017.03.108.

49. Campano, C., Merayo, N., Negro, C., et al., *Int. J. Biol. Macromol.* 114: 1077(2018b). https://doi.org/10.1016/j.ijbiomac.2018.03.170.

50. Wilson, L.A., in *What the Printer Should Know about Paper, Paper and Printability*, GATF Press, Sewickley, PA, USA, 1997, pp. 79-89.