

Advances in Nanocellulose Based Materials as Adsorbent for Wastewater Treatment

Eleen Dayana Mohamed Isa¹, Kamyar Shameli^{1,*}, Roshafima Rasit Ali¹, Vekes Balasundram¹

¹Malaysia-Japan International Institute of Technology, Universiti Teknologi Malaysia, Kuala Lumpur, Malaysia

* Correspondence: kamyarshameli@gmail.com

<https://doi.org/10.37934/jrnn.5.1.4364>

ABSTRACT

Environmental pollutions especially water pollution has been a persisting issue battle by mankind worldwide. There are various kinds of pollutants in contaminated water where it varies from heavy metals, organic dyes, pharmaceutical products and others. The presence of all these pollutants poses adverse effect to both animal and human as most of them are toxic. With the depleting sources of clean water, the needs of treatment for polluted water are highly demanded. Adsorption process for removal of contaminant has been gaining popularity due to the easy application. Nanocellulose based materials emerged as one of the most interesting adsorbents to be applied. This is due to its sustainability in production which is aligned with the current interest of research. The main source of nanocellulose is plants and nowadays, majority of researches utilize agricultural and industrial wastes as the sources. This article focuses on the nanocellulose and its classification, the sources of nanocellulose and its application as adsorbent for various types of pollutants.

Keywords:

Nanocellulose, Adsorbent,
Wastewater remediation,
Environmental pollutions

Received: 12 December 2021

Revised: 8 April 2022

Accepted: 10 April 2022

Published: 17 April 2022

1. Introduction

Water pollution is one of the biggest problems plaguing everybody around the world. It is known that clean water is essential for survival of mankind. However, with industrial revolution, it is nearly impossible to obtain clean water straight from its source. The wastewater from the industries were directly released to water bodies thus contaminate and pollute the clean water source. The types of pollutants present in contaminated water varies from heavy metals, organic

dyes, oils and solvents, pharmaceuticals and other organic compounds (Singh et al., 2020). These pollutants will pose adverse effect towards both animal and mankind. Therefore, removal technique that is non-selective is much needed. There are various methods to remove pollutants such as physical process; mainly filtration, chemical process; chemical coagulation and biological process; such as activated sludge (Gopakumar, Manna, Pasquini, Thomas, & Grohens, 2018). Although these techniques are effective to remove most of the pollutants, it high energy and cost, production of sludge and sometimes toxic by-product make it undesirable (Saharan, Pinjari, Gogate, & Pandit, 2014; Sahu & Singh, 2019). With the recent focus towards sustainable and eco-friendly materials for water purification, nanocellulose production for wastewater remediation has been gaining popularity. This is due to their desirable properties such as high surface area to volume ratio, abundant hydroxyl groups which make it easy for functionalization, good chemical resistance and good mechanical properties (Gopakumar et al., 2018). This chapter focused on nanocellulose and its classification and the potential of nanocellulose based materials as adsorbent for wastewater remediation.

2. Nanocellulose

Recently, the area of green, renewable and sustainable materials has been gaining popularity and importance for both academicians and industries as it provides an alternative solution to environmental pollution, global issues and currently depleting non-renewable resources (Trache et al., 2020). Biobased compounds such as cellulose, starch, alginate, chitin, chitosan and other polysaccharides emerged as potential alternative source in producing materials with the same properties as petroleum-based materials (Nguyen, Naficy, Chandrawati, & Dehghani, 2019). Among the polysaccharides, cellulose is the most abundant that can be found in various plants, algae, tunicates and microorganism. Statistic shows that the annual production of cellulose biomass is about 1011-1012 tons and only small portion, 6×10^9 tons, is used for industries thus makes it the most abundant raw materials (Köse, Mavlan, & Youngblood, 2020). The term "cellulose", originating from a French word "cellule" with a definition of living cell and glucose, was first discovered by a French chemist, Anselme Payen, in 1838 (Tshikovhi, Mishra, & Mishra, 2020). Cellulose is made out of hydrogen, oxygen and carbon in the form of linear homopolysaccharides of anhydro-D-glucose unit linked by β -1,4-glycosidic bond which form a cellobiose, a repeating unit of cellulose (Phanthong et al., 2018; R et al., 2021). Figure 1 shows the cellobiose of cellulose. Cellulose exist in four different polymorphs known as cellulose I, II, III and IV and it can be transformed via thermal or chemical treatments (Trache et al., 2020). Naturally, cellulose exist as assemblies of chain. Through intramolecular and intermolecular hydrogen bonding, the cellulose chains form fibrils thus make it strong and highly stable.

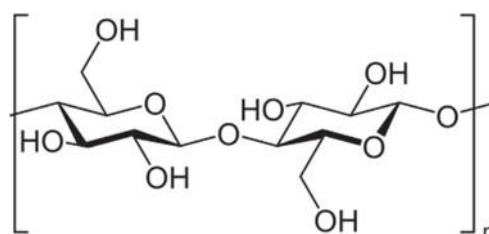


Figure 1. Chemical Structure of Cellobiose.

Besides that, this strong hydrogen bonding due to abundance of hydroxyl group leads to formation of crystal structure (Tshikovhi et al., 2020). Cellulose has been widely studied in the past decades but its nanosized form has been receiving attention these last two decades.

2.1. Classification of Nanocellulose

Nanocellulose can be defined as cellulose materials with one of its dimension having 100 nm or less (Faiz Norrrahim et al., 2021). Due to its reduction of size to nanometer, they exhibit desirable properties such as high surface area to volume ratio, good strength, low thermal expansion, capability of forming hydrogen bond, easy functionalization, biocompatible, environmentally friendly, renewability and non-toxic (Trache et al., 2020). In general, nanocellulose can be classified into three which are cellulose nanocrystals (CNCs), cellulose nanofibrils (CNFs) and bacterial nanocellulose (BNCs). There are also others slightly recent discovery of nanocellulose such as amorphous nanocellulose, cellulose nanoyarn and cellulose nanoplatelets (Chávez-Guerrero, Sepúlveda-Guzmán, Silva-Mendoza, Aguilar-Flores, & Pérez-Camacho, 2018; Grumezescu, 2016; Prado, Jacinto, & Spinace, 2020; Sen Wang, Lu, & Zhang, 2016). However, this newer classification nanocellulose is not covered in this chapter. Figure 2 shows the common classification of nanocellulose.

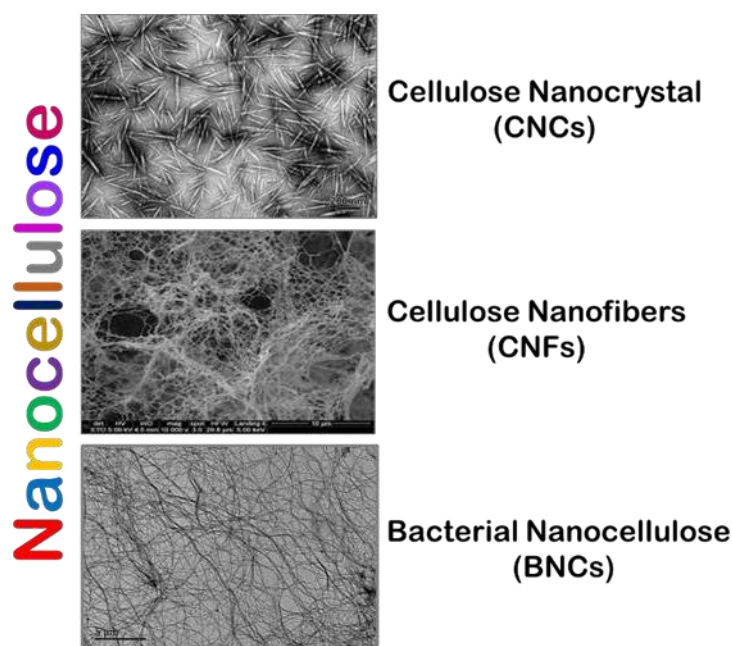


Figure 2. Classification of nanocellulose from cellulose nanocrystal, cellulose nanofiber and bacterial nanocellulose.

2.1.1. Cellulose Nanocrystals (CNCs)

Cellulose nanocrystals (CNCs) are extracted from the crystalline part of cellulose via top-down approach. There are various extraction methods such as mechanical shearing, enzymatic hydrolysis and the most common and popular is acid hydrolysis. Through the acid hydrolysis process, the amorphous region of cellulose will be hydrolyzed while the crystalline region remain intact due to its higher resistivity towards acid which then produced CNCs (Shojaeiarani, Bajwa, & Shirzadifar, 2019). CNCs dimensions can varied from 5 to 10 nm and 50 to 500 nm in width and length respectively. The properties of CNCs dependent on the source of cellulose and its crystallinity and the extraction process used (Thakur, Thakur, Raghavan, & Kessler, 2014). CNCs has high percentage of crystalline structure which makes it less flexible (Zinge & Kandasubramanian, 2020). Typically,

CNCs exist in cylindrical and rod-like morphologies but in these recent years, other shapes such as nanoballs, nanowhiskers and other were also generated (Trache et al., 2020).

2.1.2. Cellulose Nanofibrils (CNFs)

Another common class nanocellulose is cellulose nanofibrils (CNFs). CNFs consists of interconnected network of fibrils made of both crystalline and amorphous domains. The lateral size of CNFs is in the nanometer range (1-100 nm) while the length can go up to several micrometer (Kalia, Boufi, Celli, & Kango, 2014). The characteristics of CNFs is similar to CNCs in exception of lower crystallinity due to the presence of amorphous component (Ghasemlou, Daver, Ivanova, Habibi, & Adhikari, 2021). CNFs was first discovered by Turbak, Synder and Sandberg in the 1970s (Kalia et al., 2014). The extraction of CNFs greatly differs than CNCs where mechanical techniques were employed. Some of the common extraction methods of CNFs are high pressure homogenization, microfluidization, grinding and ultrasound (Ghasemlou et al., 2021; Kalia et al., 2014).

2.1.3. Bacterial Nanocellulose

Bacterial nanocellulose (BNCs) as it is named, produced using bacteria via bottom-up approach in which it is produce as exopolysaccharide of cellobiose (Phanthong et al., 2018). The production of BNCs generally proceed through four stages; (a) activation of monosaccharide via sugar nucleotide development, (b) polymerization of repeating units, (c) addition of acyl groups if present and finally (d) excretion of BNCs from the microorganism (C. Sharma & Bhardwaj, 2019). Various bacteria such as *Gluconacetobacter* (also known as *Acetobacter*), *Acanthamoeba*, *Achromobacter*, *Aerobacter*, *Agrobacterium*, *Alcaligenes*, *Azotobacter*, *Escherichia*, *Pseudomonas*, *Rhizobium*, *Salmonella*, *Sarcina* and *Zooglea*. However, the most effective species in producing BNCs is a Gram-negative bacteria, *Acetobacter xylinum* (Skočaj, 2019). Due to its production nature, BNCs is free from other components such as lignin, hemicellulose, pectin and others that can be found from lignocellulosic biomass (Phanthong et al., 2018). The BNCs produced typically highly crystalline with ribbon-shape fibrils and diameter ranging from 20-100 nm and the length can reach up to several micrometers. By controlling the experimental conditions, the size of BNCs can be altered (Ghasemlou et al., 2021).

3. Sources of Nanocellulose

Prior of obtaining nanocellulose, the cellulose must be obtained and the sources of cellulose varied from plants (inclusive of biomass and agricultural wastes), bacteria, algae and industrial wastes. Figure 3 and Table 1 shows the sources of cellulose for the production of nanocellulose.

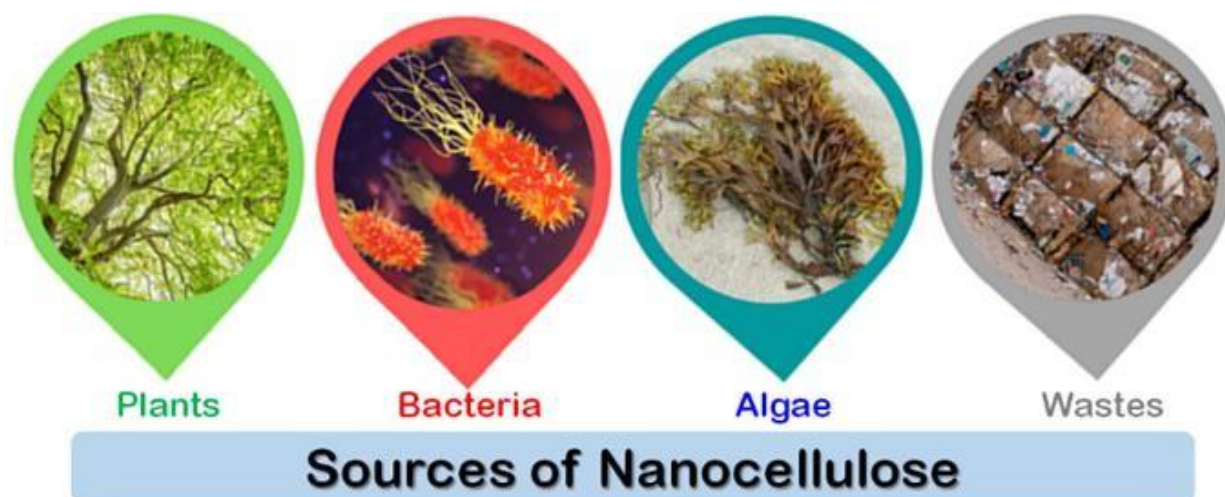


Figure 3. Sources of nanocellulose prepared from plants, bacteria, algae, and wastes.

Table 1. Sources of cellulose for production of nanocellulose.

Sources	Species/sources type	Type of nanocellulose	Ref
Plants	Natural cotton	CNCs	(Theivasanthi, Anne Christma, Toyin, Gopinath, & Ravichandran, 2018)
	<i>Zea mays</i> husk	CNCs	(Onkarappa, Prakash, Pujar, Rajith Kumar, V, et al., 2020)
	Native rubber wood powder, Maize husk, Sugar cane bagasse	CNCs	(Onkarappa, Prakash, Pujar, Kumar, et al., 2020)
	Carrot pomace	CNCs	(Cieśla, Chylińska, Zdunek, & Szymańska-Chargot, 2020)
	<i>Hevea brasiliensis</i>	CNCs	(Onkarappa, Prakash, Pujar, Rajith Kumar, Latha, et al., 2020)
	Oil palm frond leaves	CNCs	(Elias et al., 2017)
	Pomegranate peel	CNCs	(Dewan et al., 2021)
	Pineapple peel residues	CNCs	(Camacho et al., 2017)
	<i>Pyrus pyrifolia</i> fruit peel	CNCs	(Y. W. Chen, Hasanulbasori, Chiat, & Lee, 2019)
	<i>Eichhornia crassipes</i>	CNCs	(Tsade Kara, Anshebo, Sabir, & Adam Workineh, 2021)
	Rubber wood Kenaf bast fibre	CNCs	(Tuerxun et al., 2019)
	Corn cob	CNCs	(Louis & Venkatachalam, 2020)
	Waste reed	CNCs	(C. Zhang et al., 2020)
	Agricultural byproducts	CNCs	(Shahi, Wang, Adhikari, Min, & Rangari, 2021)
<ul style="list-style-type: none"> • Peanut shell • Rice husk • Sugar can bagasse 			
Bacteria	<i>Gluconacetobacter xylinus</i> strain	BNCs	(Yingkamhaeng, Intapan, & Sukyai, 2018)

	<i>Medusomyces gisevii</i> Sa-12	BNCs	(Sakovich, Skiba, Gladysheva, Golubev, & Budaeva, 2020)
	<i>Cellulomonas uda</i> <i>Clostridium cellulosi</i>	BNCs	(Zhong, Zajki-Zechmeister, & Nidetzky, 2021)
	<i>Komagataeibacter medellinensis</i> ID13488 strain	BNCs	(Jeremic et al., 2019)
	<i>Komagataeibacter hansenii</i>	BNCs	(Jacek, Ryngajłło, & Bielecki, 2019)
	<i>Komagataeibacter saccharivorans</i> MD1	BNCs	(Abol-Fotouh et al., 2020)
Algae	Cystoseria myricaas algae	CNCs	(Zarei, Niad, & Raanaei, 2018)
	<i>Gelidium elegans</i> red algae marine biomass	CNCs	(Y. W. Chen, Lee, Juan, & Phang, 2016)
	Antarctic algae <i>Cystosphaera jacquinottii</i>	CNFs	(Paniz et al., 2020)
	Cladosphora sp. algae	CNCs	(J. Liu, Willför, & Mihranyan, 2017)
	<i>Gelidium sesquipedale</i>	CNCs	(de Oliveira et al., 2019)
	<i>Enteromorpha prolifera</i>	CNFs	(Z. Zhang, Liu, Wang, He, & Bai, 2021)
	<i>Pithophora</i>	CNCs	(Gustafsson et al., 2019)
	<i>Aegagropila linnaei</i>	CNFs	(Guo et al., 2017)
	<i>Ulva lactuca</i>	CNFs	(Wahlström et al., 2020)
	<i>Laminaria hyperborea</i>	CNFs	(Onyianta, O'Rourke, Sun, Popescu, & Dorris, 2020)
Wastes	Pulp and paper mills sludge	CNCs	(Gibril et al., 2018)
	Industrial kelp (<i>Laminaria japonica</i> algae) waste	CNCs	(Z. Liu, Li, Xie, & Deng, 2017)
	Discarded cigarette filters	CNCs	(Ogundare, Moodley, & van Zyl, 2017)
	Municipal solid wastes <ul style="list-style-type: none"> • <i>Panax ginseng</i> • Spent tea residue • Waste cotton cloth • Old corrugated cardboard 	CNCs	(Y. W. Chen & Lee, 2018)
	Baby diaper wastes	CNCs	(Trilokesh, Bavadharani, Mahapriyadarshini, Janani, & Uppuluri, 2021)
	Recycled Tetra Pak Aseptic	CNCs	(Diop & Lavoie, 2017)

3.1. Plants

Plants are the most common source of cellulose in the production of nanocellulose. They are abundant, available and easily accessible. Various parts of plants can be used as the source as the plant cell wall consist of cellulose (25-50 %), hemicellulose (20-35 %) and lignin (10-25 %) (Ibrahim, Sazali, Salleh, & Ismail, 2021). However, with the recent focus towards sustainable process, by-products of plants and agriculture have been the main source of nanocellulose. The common process

of obtaining nanocellulose from plants is through acid hydrolysis via top-down approach. Onkarappa and colleagues reported the production of nanocellulose from native rubber, wood powder, maize husk and sugar cane bagasse. The cellulose component was extracted through alkali treatment and bleaching (Onkarappa, Prakash, Pujar, Kumar, et al., 2020). In another work, two step acid hydrolysis process was employed to extract nanocellulose from pineapple peel residues. The first step was using hydrochloric acid and the product obtained was microcrystalline cellulose. The size reduction to nanometer occurred through the second step acid hydrolysis using sulfuric acid (Camacho et al., 2017). Agriculture wastes such as peanut shells, rice husk and sugarcane bagasse were used the source of cellulose. The extraction process yielded 35, 39 and 41 % cellulose from peanut shell, rice husk and sugarcane bagasse respectively. CNFs were then produced through ultrasonic assisted chemical methods. Through FESEM analysis, the CNFs generated exhibited web-like, parallel and tangled morphologies for peanut shell, rice husk and sugarcane bagasse (Shahi et al., 2021). Interestingly, Dewan and co-workers reported a simple process of microwave technique in producing nanocellulose. The nanocellulose generated exhibits distorted honeycomb-like cage structure with diameter of 1-16.7 μm (Dewan et al., 2021).

3.2. Bacteria

The production of nanocellulose using bacteria proceed via bottom-up approach and the type of nanocellulose being produced is BNCs. The most commonly used bacteria in BNCs production are *Gluconacetobacter xylinus* and based on FESEM analysis, BNCs with an ultrafine network structure with diameter between 70-100 nm. Another advantage of bacteria synthesized nanocellulose is its high crystallinity and purity (Yingkamhaeng et al., 2018). One literature reported the usage of 2 bacteria, *Cellulomonas uda* and *Clostridium cellulosi* to produced BNCs with diameter of 5-7 nm and up to several micrometer in length (Zhong et al., 2021). Several of *Komagataeibacter* species were also used in generation of BNCs (Abol-Fotouh et al., 2020; Jacek et al., 2019; Jeremic et al., 2019). Although bacteria able to produce good quality nanocellulose with high purity, the conditions and media of incubation in some cases are complicated and expensive.

3.3. Algae

Algae is one of widely used raw material for various industrial sector such as food, pharmaceuticals, cosmetics and others. It is due to their components such as alginates, carrageenans, agar, etc. One component that rarely explored is cellulose and this lead to various researches in discovery and extracting cellulose from algae for applications such as production of nanocellulose (Paniz et al., 2020). Red algae, *Gelidium elegans* was used as source of nanocellulose. The production proceeds through three stages of alkalization, bleaching treatment and acid hydrolysis treatment. The imaging analysis showed that the nanocellulose exhibit average diameter and length of 21 and 547 nm respectively (Y. W. Chen et al., 2016). CNFs was successfully extracted from another type of algae, *Enteromorpha prolifera*. The CNFs has diameter ranging of 20-40 nm, length of several microns and the crystallinity index of 57.2 % (Gustafsson et al., 2019). The advantage of this work is they utilized an environmental issue which cause by this alga to produce a material that can benefit the society.

3.4. Wastes

One of the most intriguing sources of nanocellulose is industrial wastes. With the concern towards the environment and increasing amount of wastes in the world, utilizing the waste to produce something useful is beneficial and the main focus in the society nowadays. Gibril and co-workers reported on the usage of pulp and paper mills sludge to produce CNCs. The sludge were collected, cleaned and the CNCs was extracted using ammonium persulphate (Gibril et al., 2018).

Another interesting waste investigated for CNCs production was discarded cigarette filters. Acid hydrolysis with sulfuric acid technique was employed to extract the CNCs from cigarette filter wastes (Ogundare et al., 2017). Diop and colleague utilized commercial food packaging, Tetra Pak food packaging, as the source of CNCs. The TEM analysis showed that the average length and width of CNCs produced was 127-258 and 11-14 nm. By manipulating the process time, the morphology and crystallinity of CNCs can be altered (Diop & Lavoie, 2017).

4. Nanocellulose Based Materials for Wastewater Remediation

Water pollution is one of the biggest problems plaguing everybody around the world. It is known that clean water is essential for survival of mankind. However, with industrial revolution, it is nearly impossible to obtain clean water straight from its source. The wastewater from the industries were directly released to water bodies thus contaminate and pollute the clean water source. The presence of various different type of pollutants such as heavy metals, dyes, pharmaceutical compounds, oil and organic solvent, microorganism and others will pose adverse effect to both animal and mankind health (Singh et al., 2020). The removal of these toxic compounds from the wastewater and polluted water is the priority nowadays. There are various techniques that can be employed such as the conventional wastewater treatment plant which involve physical process such as filtration and adsorption, chemical process such as coagulation and catalytic degradation and biological process which uses microbes to digest the waste via both aerobic and anaerobic process. Although these techniques are effective to remove most of the pollutants, it high energy and cost, production of sludge and sometimes toxic by-product make it undesirable (Saharan et al., 2014; Sahu & Singh, 2019). Therefore, a non-selective sustainable materials and simple pollutant removal process is needed to overcome both issues of water pollution and complicated conventional treatment technique.

Nanocellulose emerged as a promising material for wastewater and polluted water remediation. This is due to their properties such as high surface area to volume ratio, abundant hydroxyl groups which make it easy for functionalization, good chemical resistance and good mechanical properties (Gopakumar et al., 2018). One of the most important factors that make it favorable over other materials is the source of nanocellulose. With the recent focus towards sustainability, nanocellulose materials are produced using wastes from agricultures, industries and even domestic. Typically, nanocellulose based materials are being used as adsorbent of various types of pollutants from water.

5. Nanocellulose Based Materials as Adsorbent

Adsorption process is the most common technique used by nanocellulose based materials for waste water remediation. This technique was recognized worldwide as the one of the most cost effective and efficient in removal of pollutants. In general, there are two adsorption mechanism which are chemisorption and physisorption. True to its name, chemisorption refers to the formation of chemical bonds between the pollutant and the surface of adsorbent. The bond can vary from ionic exchange, electrostatic interaction and chelation. Physisorption mechanism involves mainly the interaction through Van der Waals (Singh et al., 2020). In this section, we will focus on nanocellulose based materials as adsorbent for various pollutants such as heavy metals, organic dyes, oil and solvent, pharmaceuticals and other organic compounds. Table 2 shows the nanocellulose based materials as adsorbent for various different type of pollutants.

Table 2. Nanocellulose based materials as adsorbent for different pollutants.

Pollutant type	Specificities of pollutant	Nanocellulose composites	Ref
Heavy metals	Hg	Fe ₃ O ₄ -nanocellulose NCs	(Zarei et al., 2018)
		Spherical nanocellulose	(Ram & Chauhan,
		Thiol-functionalize spherical nanocellulose	2018)
		Nitro-oxidized carboxycellulose nanofibers	(H. Chen et al., 2021)
		CNF-lysozyme nanofibrils	(Silva et al., 2020)
	As	Fe ₃ O ₄ -nanocellulose NCs	(Baruah et al., 2020)
		Nanocellulose-polyethyleneimine-glutaraldehyde NCs	(Chai et al., 2020)
	Co	Sulfhydryl and carboxyl functionalized magnetite nanocellulose composites	(Anirudhan, Shainy, & Deepa, 2019)
	Cd	Nanocellulose	(P. R. Sharma et al., 2018)
	Pb	Carboxylated chitosan/carboxylated nanocellulose hydrogel beads	(Xu, Ouyang, & Yang, 2021)
		CNFs and CNCs	(Ramos-Vargas, Huirache-Acuña, Guadalupe Rutiaga-Quiñones, & Cortés-Martínez, 2020)
		CNFs/cystal nanocellulose sheets (CNS)	(Hernández-Francisco et al., 2020)
Cr	Nanocellulose and succinic anhydride functionalize nanocellulose	(Kara, Anshebo, & Sabir, 2020)	
Mn	Dithiozone modified nanocellulose	(Ahmadi-Asoori, Tazikeh-Lemeski, Mirabi, Babanezhad, & Juybari, 2021)	
Ni	CO-CNCs	(Yu, Zhang, Wang, Yin, & Huang, 2021)	
Th	Polymer grafted nanocellulose/GO NCs	(Sreenivasan, Mahesh, & Sumi, 2019)	
U	CNFs aerogel	(Y. Wang et al., 2021)	
	Phosphorylated CNFs	(Lehtonen et al., 2020)	

	V, Cr	Modified nanocellulose	(Daniel, Zahir, & Asghar, 2021)
	Pb, La	Nanocellulose scaffolds	(Zhan et al., 2020)
	Cu, Pb, Cd	CNF/chitosan/mmt NCs	(Rong et al., 2021)
	Cr, Pb, Hg	CNFs/GO/Fe ₃ O ₄	(Hosseini, Zaki Dizaji, Taghavi, & Babaei, 2020)
	Cd, Mg, Fe,	Chitosan nanocellulose	(Grzabka-Zasadzińska, Ratajczak, Król, Woźniak, & Borysiak, 2021)
	Cr, Cu, Co, Ni, Mn, Zn, Ti, Fe, Zr	MOF-CNFs	(Li, Tan, & Xu, 2020)
Organic dyes	Methylene blue	Sodium periodate-modified nanocellulose NCs	(Tsade Kara et al., 2021)
		Carboxylated CNCs	(Wu & Andrews, 2020)
		CNFs composite aerogel	(Sijie Wang, Zhang, Wang, & Pu, 2020)
		CNFs aerogel	(Beh, Lim, Lew, & Lai, 2020)
	Malachite green	GO-Nanocellulose-Copper NCs	(Khawaja, Zahir, Asghar, & Asghar, 2021)
	Victoria Blue R	Chitosan-nanocellulose Cs	(Frye, Vasisth, Atassi, Mazyck, & Nino, 2021)
	Methylene blue	Dendritic colloidal silica-hairy nanocellulose NCs	(Nia, Tavakolian, Kiasat, & van de Ven, 2020)
	Methyl orange		
	Methylene blue	Starch/Magnetite functionalized nanocellulose	(Moharrami & Motamedi, 2020)
	Crystal violet		
Direct Blue 6	Nanocellulose-polypyrrole	(Tasrin, Mohamed Madhar Fazil, Senthilmurugan, & Selvaraju, 2021)	
Bromophenol blue			
Naphthol blue black	Nanocellulose	(Riva, Pastori, Panozzo, Antonelli, & Punta, 2020)	
Orange II			
Brilliant Blue R			

	Cibacron Brilliant Yellow		
Oil and organic solvents	Thiophene Ethanol Ethyl acetate Cyclohexane Sesame oil Acetone Dichloromethane	Al ₂ O ₃ - nanocellulose NCs	(X. Zhou, Fu, Liu, Gu, & Guo, 2021)
	Mineral oil Sesame oil Ethyl acetate Thiophene Pump oil Waste pump oil Kerosene Ethanol	Nanochitosan/rGO/ Nanocellulose NCs	(Gu et al., 2021)
	N/A	Nanocellulose aerogel	(Dilamian & Noroozi, 2021)
	N/A	MOF-nanocellulose	(Li, Zhai, Wu, & Xu, 2021)
	Chloroform N/A	Nanocellulose aerogel Graphene/PVA-nanocellulose	(Shang et al., 2021) (L. Zhou & Xu, 2020)
Pharmaceuticals	Tetracycline Vancomycin Ciproflaxin, Amoxicillin Chlohexidine	Iron(III) loaded CNFs Functionalize nanocellulose	(Lu, Liu, Chen, & Luo, 2021) (Vismara et al., 2021)
	Ciprofloxacin Diclofenac	Nanocellulose	(Shahnaz, Vishnu Priyan, Pandian, & Narayanasamy, 2021)
Other organic compounds	Chlorpyrifos (Pesticide) Diuron (Herbicide) Tetraconazole (Pesticide) Fluoride	Polyvinylamine-modified nanocellulose MMT-nanocellulose Nanocellulose Nanocellulose/PVA composites	(Yang et al., 2020) (Ma, Yi, Yang, Tao, & Li, 2020) (Khalaf et al., 2021) (Das, Das, Bhowal, & Bhattacharjee, 2021)

	Nitrate	3-chloro propyl trimethoxysilane modified nanocellulose	(Manhooei, Mehdinejadani, & Amininasab, 2020)
	Nitrate Nitrite Phosphate	CNFs aerogel	(Darabitabar, Yavari, Hedayati, Zakeri, & Yousefi, 2020)
	Nitrate	3-chloro propyl trimethoxysilane modified nanocellulose	(Manhooei et al., 2020)
Two Different Types of Pollutants			
Heavy metals	Chromium	Nanocellulose-polypyrrole Cs	(Shahnaz, S, V.C, & Narayanasamy, 2020)
Organic dyes	Congo red dye		
	Lead Methylene blue	Hydroxyapatite-nanocellulose NCs	(Oun, Kamal, Farroh, Ali, & Hassan, 2021)
	Copper Malachite green dye	Polyetherimide-Carboxylated nanocellulose	(Tang et al., 2020)
	Arsenic Rhodamine B	MOFs-CNFs	(Ashour, Abdel-Magied, Wu, Olsson, & Forsberg, 2020)
Organic compounds	Cyanotoxin	CNFs grafted with beta cyclodextrin	(Gomez-Maldonado et al., 2021)
Organic dyes	Methylene blue dye		
Organic dyes	Methylene blue dye	GO-CNFs aerogel	(Z. Wang, Song, Wang, Zhang, & Yao, 2021)
Pharmaceuticals	Tetracycline		
	Methyl orange Diclofenac	MOFs-nanocellulose	(KarzarJeddi, Laitinen, Mahkam, & Liimatainen, 2020)
Organic dyes	Bromophenol blue dye	Nanobentonite-nanocellulose chitosan based aerogel	(V. Sharma, Shahnaz, Subbiah, & Narayanasamy, 2020)
Oil and solvents	Direct blue 6 dye Isopropanol Ethanol Methanol Spent engine oil Silicon oil Ethylene glycol		
Heavy metals	Copper Chloroform	CNFs aerogel	(Ji, Wen, Wang, Zhang, & Guo, 2020)

Oil and organic solvents

5.1. Heavy metals

One of the most concerning pollutants in the wastewater nowadays is heavy metals. It can be defined as any dense (density > 5 g/cm³) metal or metalloid that pose adverse effect. World Health Organization (WHO) stated that some heavy metals such as cadmium, mercury, lead and arsenic are in the top 10 chemicals of concern. The presence of heavy metals in water is resulted from natural phenomena such as weathering of soils and rocks. However, the most common contributor of heavy metals in water is the industries such as mining, metal plating, paper and pesticides. Due to its insusceptibility to degradation, it tends to accumulate in living organism. High concentration of heavy metals will cause neurological diseases, cancer, organ injury and in serious case, mortality. Therefore, it is important to remove these heavy metals from water (Gopakumar et al., 2018; R et al., 2021). Among various technique to heavy metals, adsorption technique emerged as the most popular method to be applied in heavy metals removal. With the emphasis of sustainable material nowadays, bioadsorbent such as nanocellulose gained popularity as heavy metals adsorbent. Pure nanocellulose in the form of carboxymethylcellulose nanofiber was extracted from Australian spinifex grass was used to remove cadmium from water. The results indicated that the CNFs produced able to removed cadmium from the water. At low concentration and high concentration, the nanocellulose and cadmium interacted via chelation and crystallization respectively (P. R. Sharma et al., 2018). Ram and co-workers report on the production of nanocellulose with spherical morphology for adsorption of mercury from water. Besides that, they also functionalized the spherical nanocellulose with thiol group and compared the adsorption activity between the two nanocellulose. It was found that with thiol functionalized nanocellulose, the adsorption of mercury reach up to 97 % within 20 minutes with 100 ppm solution concentration (Ram & Chauhan, 2018). Composite consist of nanocellulose, chitosan and montmorillonite were used as adsorbent for various heavy metals such as copper, lead and cadmium. An aerogel form of the composites was used and it is capable to adsorb the three different heavy metals (Rong et al., 2021). Nanocellulose based materials was also used as adsorbent for radionuclides such as thorium and uranium. Polymer grafted nanocellulose functionalized with graphene oxide was used as adsorbent for thorium and the result showed that up to 98 % of thorium was adsorb (Sreenivasan et al., 2019). CNFs aerogel was successfully produced via simple crosslinking and freeze-drying technique. The adsorption of uranium indicated that the produced CNFs aerogel has maximum adsorption capacity of 195 mg g⁻¹ with initial concentration of 10 ppm (Y. Wang et al., 2021).

5.2. Organic dyes

Dyes are defined as compounds that can adhere themselves to surfaces or fabrics to generate bright and lasting colour. Before 1856, natural dyes were used and they were obtained through extraction process from plants and animal resources (Carmen & Daniela, 2012). Although these natural dyes seem more environmentally friendly and sustainable, they actually poses several disadvantages such as the extraction process uses many hazardous chemicals, limited shade range and limited availability (Richards, 2015). In the year 1856, the first synthetic dye was discovered by W. H. Perkin which was "mauve", a brilliant bright fuchsia colour (Carmen & Daniela, 2012). With this discovery, the usage of synthetic dyes rapidly replaced the natural dyes as it overcame the drawbacks posed by natural dyes. As time moves on, more synthetic dyes were produced to be used

across various fields such as textile, cosmetic, paper, leather, pharmaceuticals, food and agriculture. In fact, up to the current date, there are over 100 000 commercial dyes available with a yearly production of over 7×10^5 tonnes per year (Brillas & Martínez-Huitle, 2015; Wee & Lim, 2016). Naturally, with increasing production, increase of waste occur. Hence, there are 450 000 ton of organic dyes produced yearly worldwide with more than 11% is lost during manufacturing and application process (Yagub, Sen, Afroze, & Ang, 2014). This leads to dye becoming the major pollutant to water. Besides that, these commercial organic dyes are made out of complex aromatic structure which makes them resistant to degradation (Drumond Chequer et al., 2013). Similarly as heavy metals, organic dyes can be removed via adsorption using nanocellulose based materials. The most common dye being studied as model textile wastewater is methylene blue. Nowadays, recent researches have been focused towards the production of nanocellulose aerogels as adsorbent due to their properties such as high surface area. CNFs aerogel was reportedly produced from sago pith via three steps of dewaxing and delignification, sonification and homogenization and finally freeze-drying. The results for removal of methylene blue indicated that the sample manage to remove up to 99 % of the dye (Beh et al., 2020). Composite consist of nanocellulose and chitosan managed to removed Victoria Blue R dye. The adsorption followed pseudo-second order kinetics and Langmuir isotherm (Frye et al., 2021). Besides that, nanocellulose based materials were also used to studied the adsorption of different dyes. Nia and colleagues produced a new type of silica-nanocellulose hybrid aerogel for adsorption of methylene blue and methyl orange. The fabricated aerogel has adsorption capacity of 270 and 300 mg/g of methylene blue and methyl orange dyes respectively.

5.3. Oil and solvents

The majority of oil pollution come from oil spill from tankers, but there are also other sources that cumulatively contribute to oil pollution. The main issue with oil pollution is its spread rapidly especially lighter oils. It also covers the surface of water which effect the aquatic ecosystem adversely and contaminate the drinking water for mankind (Gopakumar et al., 2018). Nanocellulose is of an interesting material for oil and solvents adsorption. Due to abundant hydroxyl groups on its surface, it can be functionalized to make it hydrophobic thus good adsorbent for oil and solvents. Nanocellulose aerogel was produced from rice straw via freeze-drying method and it was functionalized with methyltrimethoxysilane to make it hydrophobic. The hydrophobic nanocellulose aerogel capable to adsorb various organic solvents and oils with adsorption capacity of 170 g g^{-1} (Dilamian & Noroozi, 2021). Nanocellulose composite with nanochitosan and reduce graphene oxide successfully adsorbed mineral oil, sesame oil, acetone, ethyl acetate, thiophene, pump oil, waste pump oil, kerosene and ethanol with high adsorption capacity (Gu et al., 2021).

5.4. Pharmaceuticals

There have been a lot of concerns regarding emerging pollutants such as hormones, pesticides, pharmaceutically active compounds and personal care products. As the world population grows, the demand and consumption of such products increases and this subsequently raises their concentration level in the water due to improper disposal and effluents from municipal and industries (Sabouni & Gomaa, 2019). Among the emerging pollutants, pharmaceutical waste has the highest percentage of detection in the water compared to the other emerging pollutants. Antibiotics specifically are the major concern as their consumption are very high in both human and veterinary medicine. Their presence in the environment may lead to the development of antibiotic resistant bacteria (Elmolla & Chaudhuri, 2010; Maniakova et al., 2020).The utilization of nanocellulose based materials as

adsorbent for pharmaceutical products is rather low. Shahnaz and co-workers reported on the production of nanocellulose from *Cyperus rotundas* grass for removal of ciprofloxacin and diclofenac. The maximum adsorption capacity for ciprofloxacin and diclofenac are 227 and 192 mg g⁻¹ respectively. In another research, nanocellulose was first functionalize prior to application for vancomycin, ciprofloxacin, amoxicillin and chlorhexidine adsorption. Based on the adsorption results, the adsorption process followed pseudo-second order model (Vismara et al., 2021).

5.5. Other organic compounds

Besides heavy metals, organic dyes, oil and solvents and pharmaceuticals, there are other organic compounds such as fertilizers, herbicides and pesticides were investigated as pollutants. By-products of fertilizers such as nitrate, nitrite and phosphate were adsorbed using CNFs aerogel. The overall removal percentage obtained were 79, 73 and 98 % for nitrate, nitrite and phosphate respectively (Darabitabar et al., 2020). In another work, the researchers studied the adsorption of fluoride by using polyvinyl alcohol (PVA) – nanocellulose composites. The results indicated that adsorption capacity of the adsorbent was 11.36 mg g⁻¹ (Das et al., 2021). The adsorption of pesticide, chlorpyrifos, was studied using polyvinylamine-modified nanocellulose. The overall removal percentage obtained was 93 % (Yang et al., 2020).

5.6. Combination of pollutants

The study of nanocellulose as adsorbent is not limited to a single type of pollutants. It is known that in polluted water, there might be different type of pollutants are present. Therefore, determining that whether the nanocellulose is a non-selective adsorbent is an importance in these recent years. Shahnaz and colleagues produced nanocellulose-polypyrrole nanocomposites as adsorbent of heavy metal, chromium, and organic dye, Congo red. They conducted optimization of adsorption process using response surface methodology and found that total of maximum 80 and 85 % removal percentage for chromium and Congo red respectively were obtained (Shahnaz et al., 2020). In another research, hydroxyapatite-nanocellulose composite was used to adsorb lead and methylene blue. In the absence of hydroxyapatite, the adsorption of pollutant in lower compared to the composite. Furthermore, pure nanocellulose was found to be more selective in removal of lead while composite showed excellent adsorptive capability for both pollutants (Oun et al., 2021). Arsenic and rhodamine B dye successfully removed from solution using metal-organic-framework (MOF) functionalized CNFs. The adsorption capacities of 2.8 and 2.7 mg g⁻¹ was obtained for arsenic and rhodamine B removal (Ashour et al., 2020). Besides combination of heavy metals and organic dyes, removal of combination of organic compound and dye were studied. CNFs grafted with beta cyclodextrin was produced as adsorbent for cyanotoxin and methylene blue. The adsorption capacities for both pollutants were determined to be 0.078 and 3.48 mg g⁻¹ for cyanotoxin and methylene blue respectively. There were also reports on the removal of organic dye and pharmaceutical (KarzarJeddi et al., 2020; Z. Wang et al., 2021), organic dyes and oil (V. Sharma et al., 2020) and heavy metals and oil combination (Ji et al., 2020).

4. Conclusions

Increase of world populations with time cause depletion of non-renewable sources and increment of pollutions due to the disposal of waste. Due to this, researches in the field of science, engineering and technology have been shifted towards more sustainable materials. Nanocellulose emerged as one of the most popular sustainable materials to be developed. The production of

nanocellulose can be considered as a green process as it utilized agricultural and industrial wastes. Although the extraction process might be toxic as it uses acids, further researches have proven that the process can be improved to reduce the toxicity of chemicals used. With the increasing issues related to water pollution, nanocellulose based materials has been gaining popularity as bio-adsorbent for removal of various of type of pollutants such as heavy metals, organic dyes, oil and solvents, pharmaceuticals and other organic compounds. Although the result is promising, the potential of nanocellulose based materials towards other applications especially in terms of wastewater remediation was not covered in this article.

Acknowledgement

This research was funded by Takasago Thermal Engineering Co. Ltd. grant (R.K.13000 0.7343.4B422) from the research management center (RMC) of Universiti Teknologi Malaysia (UTM) and Malaysia Japan International Institute of Technology (MJIIT).

References

- Abol-Fotouh, D., Hassan, M. A., Shokry, H., Roig, A., Azab, M. S., & Kashyout, A. E.-H. B. (2020). "Bacterial nanocellulose from agro-industrial wastes: low-cost and enhanced production by *Komagataeibacter saccharivorans* MD1." *Scientific Reports*, 10(1): 3491.
- Ahmadi-Asoori, S., Tazikheh-Lemeski, E., Mirabi, A., Babanezhad, E., & Juybari, M. H. (2021). "Preparation of nanocellulose modified with dithizone for separation, extraction and determination of trace amounts of manganese ions in industrial wastewater samples." *Microchemical Journal*, 160: 105737.
- Anirudhan, T. S., Shainy, F., & Deepa, J. R. (2019). "Effective removal of Cobalt(II) ions from aqueous solutions and nuclear industry wastewater using sulfhydryl and carboxyl functionalised magnetite nanocellulose composite: batch adsorption studies." *Chemistry and Ecology*, 35(3): 235-255.
- Ashour, R. M., Abdel-Magied, A. F., Wu, Q., Olsson, R. T., & Forsberg, K. (2020). "Green Synthesis of Metal-Organic Framework Bacterial Cellulose Nanocomposites for Separation Applications." *Polymers*, 12(5): 1104.
- Baruah, J., Chaliha, C., Kalita, E., Nath, B. K., Field, R. A., & Deb, P. (2020). "Modelling and optimization of factors influencing adsorptive performance of agrowaste-derived Nanocellulose Iron Oxide Nanobiocomposites during remediation of Arsenic contaminated groundwater." *International Journal of Biological Macromolecules*, 164: 53-65.
- Beh, J. H., Lim, T. H., Lew, J. H., & Lai, J. C. (2020). "Cellulose nanofibril-based aerogel derived from sago pith waste and its application on methylene blue removal." *International Journal of Biological Macromolecules*, 160: 836-845.
- Brillas, E., & Martínez-Huitle, C. A. (2015). "Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. An updated review." *Applied Catalysis B: Environmental*, 166-167: 603-643.
- Camacho, M., Ureña, Y.-R.-C., Lopretti, M., Carballo, L.-B., Moreno, G., Alfaro, B., & Baudrit, J.-R.-V. (2017). "Synthesis and Characterization of Nanocrystalline Cellulose Derived from Pineapple Peel Residues." *Journal of Renewable Materials*, 5(5): 271-279.
- Carmen, Z., & Daniela, S. (2012). *Textile Organic Dyes – Characteristics, Polluting Effects and Separation/Elimination Procedures from Industrial Effluents – A Critical Overview*. In T. Puzyn & A. Mostrag-Szlichtyng (Eds.), *Organic Pollutants Ten Years After the Stockholm Convention*. Rijeka: IntechOpen.
- Chai, F., Wang, R., Yan, L., Li, G., Cai, Y., & Xi, C. (2020). "Facile fabrication of pH-sensitive nanoparticles based on nanocellulose for fast and efficient As(V) removal." *Carbohydrate Polymers*, 245: 116511.
- Chávez-Guerrero, L., Sepúlveda-Guzmán, S., Silva-Mendoza, J., Aguilar-Flores, C., & Pérez-Camacho, O. (2018). "Eco-friendly isolation of cellulose nanoplatelets through oxidation under mild conditions." *Carbohydrate Polymers*, 181: 642-649.

- Chen, H., Sharma, S. K., Sharma, P. R., Chi, K., Fung, E., Aubrecht, K., . . . Hsiao, B. S. (2021). "Nitro-oxidized carboxycellulose nanofibers from moringa plant: effective bioadsorbent for mercury removal." *Cellulose*, 28(13): 8611-8628.
- Chen, Y. W., Hasanulbasori, M. A., Chiat, P. F., & Lee, H. V. (2019). "Pyrus pyrifolia fruit peel as sustainable source for spherical and porous network based nanocellulose synthesis via one-pot hydrolysis system." *International Journal of Biological Macromolecules*, 123: 1305-1319.
- Chen, Y. W., & Lee, H. V. (2018). "Revalorization of selected municipal solid wastes as new precursors of "green" nanocellulose via a novel one-pot isolation system: A source perspective." *International Journal of Biological Macromolecules*, 107: 78-92.
- Chen, Y. W., Lee, H. V., Juan, J. C., & Phang, S.-M. (2016). "Production of new cellulose nanomaterial from red algae marine biomass *Gelidium elegans*." *Carbohydrate Polymers*, 151: 1210-1219.
- Cieřła, J., Chylińska, M., Zdunek, A., & Szymańska-Chargot, M. (2020). "Effect of different conditions of synthesis on properties of silver nanoparticles stabilized by nanocellulose from carrot pomace." *Carbohydrate Polymers*, 245: 116513.
- Daniel, A. B., Zahir, E., & Asghar, M. A. (2021). "Remediation of Vanadium (V) and Chromium (III) Ions from Aqueous Media by Modified Nanocellulose Obtained from Coconut Coir." *Journal of Macromolecular Science, Part B*, 60(7): 500-520.
- Darabitar, F., Yavari, V., Hedayati, A., Zakeri, M., & Yousefi, H. (2020). "Novel cellulose nanofiber aerogel for aquaculture wastewater treatment." *Environmental Technology & Innovation*, 18: 100786.
- Das, L., Das, P., Bhowal, A., & Bhattacharjee, C. (2021). "Enhanced biosorption of fluoride by extracted nanocellulose/polyvinyl alcohol composite in batch and fixed-bed system: ANN analysis and numerical modeling." *Environmental Science and Pollution Research International*, 28(34): 47107-47125.
- de Oliveira, J. P., Bruni, G. P., Fabra, M. J., da Rosa Zavareze, E., López-Rubio, A., & Martínez-Sanz, M. (2019). "Development of food packaging bioactive aerogels through the valorization of *Gelidium sesquipedale* seaweed." *Food Hydrocolloids*, 89: 337-350.
- Dewan, A., Sarmah, M., Bharali, P., Thakur, A. J., Boruah, P. K., Das, M. R., & Bora, U. (2021). "Pd Nanoparticles-Loaded Honeycomb-Structured Bio-nanocellulose as a Heterogeneous Catalyst for Heteroaryl Cross-Coupling Reaction." *ACS Sustainable Chemistry & Engineering*, 9(2): 954-966.
- Dilamian, M., & Noroozi, B. (2021). "Rice straw agri-waste for water pollutant adsorption: Relevant mesoporous super hydrophobic cellulose aerogel." *Carbohydrate Polymers*, 251: 117016.
- Diop, C. I. K., & Lavoie, J.-M. (2017). "Isolation of Nanocrystalline Cellulose: A Technological Route for Valorizing Recycled Tetra Pak Aseptic Multilayered Food Packaging Wastes." *Waste and Biomass Valorization*, 8(1): 41-56.
- Drumond Chequer, F. M., de Oliveira, G. A. R., Anastacio Ferraz, E. R., Carvalho, J., Boldrin Zanoni, M. V., & de Oliveir, D. P. (2013). *Textile Dyes: Dyeing Process and Environmental Impact*. In *Eco-Friendly Textile Dyeing and Finishing*.
- Elias, N., Chandren, S., Attan, N., Mahat, N. A., Razak, F. I. A., Jamalis, J., & Wahab, R. A. (2017). "Structure and properties of oil palm-based nanocellulose reinforced chitosan nanocomposite for efficient synthesis of butyl butyrate." *Carbohydrate Polymers*, 176: 281-292.
- Elmolla, E. S., & Chaudhuri, M. (2010). "Comparison of different advanced oxidation processes for treatment of antibiotic aqueous solution." *Desalination*, 256(1): 43-47.
- Faiz Norrrahim, M. N., Mohd Kasim, N. A., Knight, V. F., Mohamad Misenan, M. S., Janudin, N., Ahmad Shah, N. A., . . . Zin Wan Yunus, W. M. (2021). "Nanocellulose: a bioadsorbent for chemical contaminant remediation." *RSC Advances*, 11(13): 7347-7368.
- Frye, M., Vasisth, S. E., Atassi, A., Mazyck, D., & Nino, J. C. (2021). "Effect of degree of sulfonation in nanocellulose/chitosan composite on adsorption of cationic dye as opioid simulant." *Cellulose Chemistry and Technology*, 55(1-2): 87-99.
- Ghasemlou, M., Daver, F., Ivanova, E. P., Habibi, Y., & Adhikari, B. (2021). "Surface modifications of nanocellulose: From synthesis to high-performance nanocomposites." *Progress in Polymer Science*, 119: 101418.

- Gibril, M. E., Lekha, P., Andrew, J., Sithole, B., Tesfaye, T., & Ramjugernath, D. (2018). "Beneficiation of pulp and paper mill sludge: production and characterisation of functionalised crystalline nanocellulose." *Clean Technologies and Environmental Policy*, 20(8): 1835-1845.
- Gomez-Maldonado, D., Reynolds, A. M., Johansson, L.-S., Burnett, D. J., Ramapuram, J. B., Waters, M. N., . . . Peresin, M. S. (2021). "Fabrication of aerogels from cellulose nanofibril grafted with β -cyclodextrin for capture of water pollutants." *Journal of Porous Materials*, 28(6): 1725-1736.
- Gopakumar, D. A., Manna, S., Pasquini, D., Thomas, S., & Grohens, Y. (2018). Nanocellulose: Extraction and application as a sustainable material for wastewater purification. In *New Polymer Nanocomposites for Environmental Remediation* (pp. 469-486).
- Grumezescu, A. M. (2016). Fabrication and self-assembly of nanobiomaterials: applications of nanobiomaterials. In A. M. Grumezescu (Ed.), *Fabrication and Self-Assembly of Nanobiomaterials* (pp. xxiii-xxvii): William Andrew Publishing.
- Grzabka-Zasadzińska, A., Ratajczak, I., Król, K., Woźniak, M., & Borysiak, S. (2021). "The influence of crystalline structure of cellulose in chitosan-based biocomposites on removal of Ca(II), Mg(II), Fe(III) ion in aqueous solutions." *Cellulose*, 28(9): 5745-5759.
- Gu, H., Gao, C., Zhou, X., Du, A., Naik, N., & Guo, Z. (2021). "Nanocellulose nanocomposite aerogel towards efficient oil and organic solvent adsorption." *Advanced Composites and Hybrid Materials*.
- Guo, J., Uddin, K. M. A., Mihhels, K., Fang, W., Laaksonen, P., Zhu, J. Y., & Rojas, O. J. (2017). "Contribution of Residual Proteins to the Thermomechanical Performance of Cellulosic Nanofibrils Isolated from Green Macroalgae." *ACS Sustainable Chemistry & Engineering*, 5(8): 6978-6985.
- Gustafsson, O., Manukyan, L., Gustafsson, S., Tummala, G. K., Zaman, S., Begum, A., . . . Mihranyan, A. (2019). "Scalable and Sustainable Total Pathogen Removal Filter Paper for Point-of-Use Drinking Water Purification in Bangladesh." *ACS Sustainable Chemistry & Engineering*, 7(17): 14373-14383.
- Hernández-Francisco, E., Bonilla-Cruz, J., Márquez-Lamas, U., Suárez-Jacobo, Á., Longoria-Rodríguez, F., Rivera-Haro, J., . . . Lara-Ceniceros, T. E. (2020). "Entangled cellulose nanofibrils/nanosheets derived from native mexican agave for lead(II) ion removal." *Cellulose*, 27(15): 8785-8798.
- Hosseini, M., Zaki Dizaji, H., Taghavi, M., & Babaei, A. A. (2020). "Preparation of ultra-lightweight and surface-tailored cellulose nanofibril composite cryogels derived from Date palm waste as powerful and low-cost heavy metals adsorbent to treat aqueous medium." *Industrial Crops and Products*, 154: 112696.
- Ibrahim, H., Szali, N., Salleh, W. N. W., & Ismail, A. F. (2021). "Nanocellulose-Based Materials and Recent Application for Heavy Metal Removal." *Water, Air, & Soil Pollution*, 232(7).
- Jacek, P., Ryngajłło, M., & Bielecki, S. (2019). "Structural changes of bacterial nanocellulose pellicles induced by genetic modification of *Komagataeibacter hansenii* ATCC 23769." *Applied Microbiology and Biotechnology*, 103(13): 5339-5353.
- Jeremic, S., Djokic, L., Ajdačić, V., Božinović, N., Pavlovic, V., Manojlović, D. D., . . . Nikodinovic-Runic, J. (2019). "Production of bacterial nanocellulose (BNC) and its application as a solid support in transition metal catalysed cross-coupling reactions." *International Journal of Biological Macromolecules*, 129: 351-360.
- Ji, Y., Wen, Y., Wang, Z., Zhang, S., & Guo, M. (2020). "Eco-friendly fabrication of a cost-effective cellulose nanofiber-based aerogel for multifunctional applications in Cu(II) and organic pollutants removal." *Journal of Cleaner Production*, 255: 120276.
- Kalia, S., Boufi, S., Celli, A., & Kango, S. (2014). "Nanofibrillated cellulose: surface modification and potential applications." *Colloid and Polymer Science*, 292(1): 5-31.
- Kara, H. T., Anshebo, S. T., & Sabir, F. K. (2020). "A novel modified cellulose nanomaterials (CNMs) for remediation of chromium (VI) ions from wastewater." *Materials Research Express*, 7(11): 115008.
- KarzarJeddi, M., Laitinen, O., Mahkam, M., & Liimatainen, H. (2020). "Zwitterionic hybrid aerobeads of binary metal organic frameworks and cellulose nanofibers for removal anionic pollutants." *Materials & Design*, 196: 109106.
- Khalaf, B., Hamed, O., Jodeh, S., Hanbali, G., Bol, R., Dagdag, O., & Samhan, S. (2021). "Novel, Environment-Friendly Cellulose-Based Derivatives for Tetraconazole Removal from Aqueous Solution." *Polymers*, 13(3): 450.

- Khawaja, H., Zahir, E., Asghar, M. A., & Asghar, M. A. (2021). "Graphene oxide decorated with cellulose and copper nanoparticle as an efficient adsorbent for the removal of malachite green." *International Journal of Biological Macromolecules*, 167: 23-34.
- Köse, K., Mavlan, M., & Youngblood, J. P. (2020). "Applications and impact of nanocellulose based adsorbents." *Cellulose*, 27(6): 2967-2990.
- Lehtonen, J., Hassinen, J., Kumar, A. A., Johansson, L.-S., Mäenpää, R., Pahimanolis, N., . . . Rojas, O. J. (2020). "Phosphorylated cellulose nanofibers exhibit exceptional capacity for uranium capture." *Cellulose*, 27(18): 10719-10732.
- Li, J., Tan, S., & Xu, Z. (2020). "Anisotropic Nanocellulose Aerogel Loaded with Modified UiO-66 as Efficient Adsorbent for Heavy Metal Ions Removal." *Nanomaterials*, 10(6): 1114.
- Li, J., Zhai, S., Wu, W., & Xu, Z. (2021). "Hydrophobic nanocellulose aerogels with high loading of metal-organic framework particles as floating and reusable oil absorbents." *Frontiers of Chemical Science and Engineering*, 15(5): 1158-1168.
- Liu, J., Willför, S., & Mihranyan, A. (2017). "On importance of impurities, potential leachables and extractables in algal nanocellulose for biomedical use." *Carbohydrate Polymers*, 172: 11-19.
- Liu, Z., Li, X., Xie, W., & Deng, H. (2017). "Extraction, isolation and characterization of nanocrystalline cellulose from industrial kelp (*Laminaria japonica*) waste." *Carbohydrate Polymers*, 173: 353-359.
- Louis, A. C. F., & Venkatachalam, S. (2020). "Energy efficient process for valorization of corn cob as a source for nanocrystalline cellulose and hemicellulose production." *International Journal of Biological Macromolecules*, 163: 260-269.
- Lu, L., Liu, M., Chen, Y., & Luo, Y. (2021). "Effective removal of tetracycline antibiotics from wastewater using practically applicable iron(III)-loaded cellulose nanofibres." *Royal Society Open Science*, 8(8): 210336.
- Ma, C., Yi, L., Yang, J., Tao, J., & Li, J. (2020). "Nanocellulose–organic montmorillonite nanocomposite adsorbent for diuron removal from aqueous solution: optimization using response surface methodology." *RSC Advances*, 10(51): 30734-30745.
- Manhooei, L., Mehdinejadi, B., & Amininasab, S. M. (2020). "Cellulose nanocrystal modified with 3-chloro propyl trimethoxysilane: a new bio-adsorbent for nitrate removal from water." *Water and Environment Journal*, 34(S1): 50-60.
- Maniakova, G., Kowalska, K., Murgolo, S., Mascolo, G., Libralato, G., Lofrano, G., . . . Rizzo, L. (2020). "Comparison between heterogeneous and homogeneous solar driven advanced oxidation processes for urban wastewater treatment: Pharmaceuticals removal and toxicity." *Separation and Purification Technology*, 236.
- Moharrami, P., & Motamedi, E. (2020). "Application of cellulose nanocrystals prepared from agricultural wastes for synthesis of starch-based hydrogel nanocomposites: Efficient and selective nanoadsorbent for removal of cationic dyes from water." *Bioresource Technology*, 313: 123661.
- Nguyen, L. H., Naficy, S., Chandrawati, R., & Dehghani, F. (2019). "Nanocellulose for Sensing Applications." *Advanced Materials Interfaces*, 6(18).
- Nia, M. H., Tavakolian, M., Kiasat, A. R., & van de Ven, T. G. M. (2020). "Hybrid Aerogel Nanocomposite of Dendritic Colloidal Silica and Hairy Nanocellulose: an Effective Dye Adsorbent." *Langmuir*, 36(40): 11963-11974.
- Ogundare, S. A., Moodley, V., & van Zyl, W. E. (2017). "Nanocrystalline cellulose isolated from discarded cigarette filters." *Carbohydrate Polymers*, 175: 273-281.
- Onkarappa, H. S., Prakash, G. K., Pujar, G. H., Kumar, C. R. R., Radha, V., Latha, M. S., & Betageri, V. S. (2020). "Synthesis and characterization of nanocellulose using renewable resources through Ionic liquid medium." *Advances in Natural Sciences-Nanoscience and Nanotechnology*, 11(3).
- Onkarappa, H. S., Prakash, G. K., Pujar, G. H., Rajith Kumar, C. R., Latha, M. S., & Betageri, V. S. (2020). "Hevea brasiliensis mediated synthesis of nanocellulose: Effect of preparation methods on morphology and properties." *International Journal of Biological Macromolecules*, 160: 1021-1028.
- Onkarappa, H. S., Prakash, G. K., Pujar, G. H., Rajith Kumar, C. R., V. R., & Betageri, V. S. (2020). "Facile synthesis and characterization of nanocellulose from *Zea mays* husk." *Polymer Composites*, 41(8): 3153-3159.

- Onyianta, A. J., O'Rourke, D., Sun, D., Popescu, C.-M., & Dorris, M. (2020). "High aspect ratio cellulose nanofibrils from macroalgae *Laminaria hyperborea* cellulose extract via a zero-waste low energy process." *Cellulose*, 27(14): 7997-8010.
- Oun, A. A., Kamal, K. H., Farroh, K., Ali, E. F., & Hassan, M. A. (2021). "Development of fast and high-efficiency sponge-gourd fibers (*Luffa cylindrica*)/hydroxyapatite composites for removal of lead and methylene blue." *Arabian Journal of Chemistry*, 14(8): 103281.
- Paniz, O. G., Pereira, C. M. P., Pacheco, B. S., Wolke, S. I., Maron, G. K., Mansilla, A., . . . Carreño, N. L. V. (2020). "Cellulosic material obtained from Antarctic algae biomass." *Cellulose*, 27(1): 113-126.
- Phanthong, P., Reubroycharoen, P., Hao, X., Xu, G., Abudula, A., & Guan, G. (2018). "Nanocellulose: Extraction and application." *Carbon Resources Conversion*, 1(1): 32-43.
- Prado, K. S., Jacinto, A. A., & Spinace, M. A. S. (2020). Cellulose Nanostructures Extracted from Pineapple Fibres. In M. Jawaid, M. Asim, P. M. Tahir, & M. Nasir (Eds.), *Pineapple Leaf Fibers: Processing, Properties and Applications* (pp. 185-234).
- R, R., Thomas, D., Philip, E., Paul, S. A., Madhavan, A., Sindhu, R., . . . Pandey, A. (2021). "Potential of nanocellulose for wastewater treatment." *Chemosphere*, 281: 130738.
- Ram, B., & Chauhan, G. S. (2018). "New spherical nanocellulose and thiol-based adsorbent for rapid and selective removal of mercuric ions." *Chemical Engineering Journal*, 331: 587-596.
- Ramos-Vargas, S., Huirache-Acuña, R., Guadalupe Rutiaga-Quiñones, J., & Cortés-Martínez, R. (2020). "Effective lead removal from aqueous solutions using cellulose nanofibers obtained from water hyacinth." *Water Supply*, 20(7): 2715-2736.
- Richards, P. R. (2015). Chapter 19 - Fabric Finishing: Dyeing and Colouring. In R. Sinclair (Ed.), *Textiles and Fashion* (pp. 475-505): Woodhead Publishing.
- Riva, L., Pastori, N., Panozzo, A., Antonelli, M., & Punta, C. (2020). "Nanostructured Cellulose-Based Sorbent Materials for Water Decontamination from Organic Dyes." *Nanomaterials*, 10(8): 1570.
- Rong, N., Chen, C., Ouyang, K., Zhang, K., Wang, X., & Xu, Z. (2021). "Adsorption characteristics of directional cellulose nanofiber/chitosan/montmorillonite aerogel as adsorbent for wastewater treatment." *Separation and Purification Technology*, 274: 119120.
- Sabouni, R., & Gomaa, H. (2019). "Photocatalytic degradation of pharmaceutical micro-pollutants using ZnO." *Environmental Science and Pollution Research International*, 26(6): 5372-5380.
- Saharan, V. K., Pinjari, D. V., Gogate, P. R., & Pandit, A. B. (2014). Advanced Oxidation Technologies for Wastewater Treatment. In *Industrial Wastewater Treatment, Recycling and Reuse* (pp. 141-191).
- Sahu, O., & Singh, N. (2019). Significance of bioadsorption process on textile industry wastewater. In *The Impact and Prospects of Green Chemistry for Textile Technology* (pp. 367-416).
- Sakovich, G. V., Skiba, E. A., Gladysheva, E. K., Golubev, D. S., & Budaeva, V. V. (2020). "Miscanthus as a Feedstock for the Production of Bacterial Nanocellulose." *Doklady Chemistry*, 495(2): 205-208.
- Shahi, N., Wang, P., Adhikari, S., Min, B., & Rangari, V. K. (2021). "Biopolymers Fractionation and Synthesis of Nanocellulose/Silica Nanoparticles from Agricultural Byproducts." *ACS Sustainable Chemistry & Engineering*, 9(18): 6284-6295.
- Shahnaz, T., S, M. M. F., V.C, P., & Narayanasamy, S. (2020). "Surface modification of nanocellulose using polypyrrole for the adsorptive removal of Congo red dye and chromium in binary mixture." *International Journal of Biological Macromolecules*, 151: 322-332.
- Shahnaz, T., Vishnu Priyan, V., Pandian, S., & Narayanasamy, S. (2021). "Use of Nanocellulose extracted from grass for adsorption abatement of Ciprofloxacin and Diclofenac removal with phyto, and fish toxicity studies." *Environmental Pollution*, 268: 115494.
- Shang, Q., Chen, J., Hu, Y., Yang, X., Hu, L., Liu, C., . . . Zhou, Y. (2021). "Facile Fabrication of Superhydrophobic Cross-Linked Nanocellulose Aerogels for Oil-Water Separation." *Polymers*, 13(4): 625.
- Sharma, C., & Bhardwaj, N. K. (2019). "Bacterial nanocellulose: Present status, biomedical applications and future perspectives." *Materials Science and Engineering: C*, 104: 109963.
- Sharma, P. R., Chattopadhyay, A., Sharma, S. K., Geng, L., Amiralian, N., Martin, D., & Hsiao, B. S. (2018). "Nanocellulose from *Spinifex* as an Effective Adsorbent to Remove Cadmium(II) from Water." *ACS Sustainable Chemistry & Engineering*, 6(3): 3279-3290.

- Sharma, V., Shahnaz, T., Subbiah, S., & Narayanasamy, S. (2020). "New Insights into the Remediation of Water Pollutants using Nanobentonite Incorporated Nanocellulose Chitosan Based Aerogel." *Journal of Polymers and the Environment*, 28(7): 2008-2019.
- Shojaeiarani, J., Bajwa, D., & Shirzadifar, A. (2019). "A review on cellulose nanocrystals as promising biocompounds for the synthesis of nanocomposite hydrogels." *Carbohydrate Polymers*, 216: 247-259.
- Silva, N. H. C. S., Figueira, P., Fabre, E., Pinto, R. J. B., Pereira, M. E., Silvestre, A. J. D., . . . Freire, C. S. R. (2020). "Dual nanofibrillar-based bio-sorbent films composed of nanocellulose and lysozyme nanofibrils for mercury removal from spring waters." *Carbohydrate Polymers*, 238: 116210.
- Singh, S., Kumar, V., Datta, S., Dhanjal, D. S., Sharma, K., Samuel, J., & Singh, J. (2020). "Current advancement and future prospect of biosorbents for bioremediation." *Science of the Total Environment*, 709: 135895.
- Skočaj, M. (2019). "Bacterial nanocellulose in papermaking." *Cellulose*, 26(11): 6477-6488.
- Sreenivasan, R., Mahesh, S. S., & Sumi, V. S. (2019). "Synthesis and application of polymer-grafted nanocellulose/graphene oxide nano composite for the selective recovery of radionuclides from aqueous media." *Separation Science and Technology*, 54(9): 1453-1468.
- Tang, F., Yu, H., Yassin Hussain Abdalkarim, S., Sun, J., Fan, X., Li, Y., . . . Chiu Tam, K. (2020). "Green acid-free hydrolysis of wasted pomelo peel to produce carboxylated cellulose nanofibers with super absorption/flocculation ability for environmental remediation materials." *Chemical Engineering Journal*, 395: 125070.
- Tasrin, S., Mohamed Madhar Fazil, S., Senthilmurugan, S., & Selvaraju, N. (2021). "Facile preparation of nanocellulose embedded polypyrrole for dye removal: unary and binary process optimization and seed toxicity." *International Journal of Environmental Science and Technology*, 18(2): 365-378.
- Thakur, V. K., Thakur, M. K., Raghavan, P., & Kessler, M. R. (2014). "Progress in Green Polymer Composites from Lignin for Multifunctional Applications: A Review." *ACS Sustainable Chemistry & Engineering*, 2(5): 1072-1092.
- Theivasanthi, T., Anne Christma, F. L., Toyin, A. J., Gopinath, S. C. B., & Ravichandran, R. (2018). "Synthesis and characterization of cotton fiber-based nanocellulose." *International Journal of Biological Macromolecules*, 109: 832-836.
- Trache, D., Tarchoun, A. F., Derradji, M., Hamidon, T. S., Masruchin, N., Brosse, N., & Hussin, M. H. (2020). "Nanocellulose: From Fundamentals to Advanced Applications." *Front Chem*, 8: 392.
- Trilokesh, C., Bavadarani, P., Mahapriyadarshini, M., Janani, R., & Uppuluri, K. B. (2021). "Recycling Baby Diaper Waste into Cellulose and Nanocellulose." *Waste and Biomass Valorization*, 12(8): 4299-4306.
- Tsade Kara, H., Anshebo, S. T., Sabir, F. K., & Adam Workineh, G. (2021). "Removal of Methylene Blue Dye from Wastewater Using Periodiated Modified Nanocellulose." *International Journal of Chemical Engineering*, 2021: 9965452.
- Tshikovhi, A., Mishra, S. B., & Mishra, A. K. (2020). "Nanocellulose-based composites for the removal of contaminants from wastewater." *International Journal of Biological Macromolecules*, 152: 616-632.
- Tuerxun, D., Pulingam, T., Nordin, N. I., Chen, Y. W., Kamaldin, J. B., Julkapli, N. B. M., . . . Johan, M. R. B. (2019). "Synthesis, characterization and cytotoxicity studies of nanocrystalline cellulose from the production waste of rubber-wood and kenaf-bast fibers." *European Polymer Journal*, 116: 352-360.
- Vismara, E., Bertolini, G., Bongio, C., Massironi, N., Zarattini, M., Nanni, D., . . . Torri, G. (2021). "Nanocellulose from Cotton Waste and Its Glycidyl Methacrylate Grafting and Allylation: Synthesis, Characterization and Adsorption Properties." *Nanomaterials*, 11(2): 476.
- Wahlström, N., Edlund, U., Pavia, H., Toth, G., Jaworski, A., Pell, A. J., . . . Richter-Dahlfors, A. (2020). "Cellulose from the green macroalgae *Ulva lactuca*: isolation, characterization, optotracing, and production of cellulose nanofibrils." *Cellulose*, 27(7): 3707-3725.
- Wang, S., Lu, A., & Zhang, L. (2016). "Recent advances in regenerated cellulose materials." *Progress in Polymer Science*, 53: 169-206.
- Wang, S., Zhang, Q., Wang, Z., & Pu, J. (2020). "Facile fabrication of an effective nanocellulose-based aerogel and removal of methylene blue from aqueous system." *Journal of Water Process Engineering*, 37: 101511.
- Wang, Y., Li, Y., Zhang, Y., Zhang, Z., Li, Y., & Li, W. (2021). "Nanocellulose aerogel for highly efficient adsorption of uranium (VI) from aqueous solution." *Carbohydrate Polymers*, 267: 118233.

- Wang, Z., Song, L., Wang, Y., Zhang, X.-F., & Yao, J. (2021). "Construction of a hybrid graphene oxide/nanofibrillated cellulose aerogel used for the efficient removal of methylene blue and tetracycline." *Journal of Physics and Chemistry of Solids*, 150: 109839.
- Wee, S., & Lim, C. K. (2016). "Removal of dye by adsorption: A review." 11: 2675-2679.
- Wu, J., & Andrews, M. P. (2020). "Carboxylated Cellulose Nanocrystal Microbeads for Removal of Organic Dyes from Wastewater: Effects of Kinetics and Diffusion on Binding and Release." *ACS Applied Nano Materials*, 3(11): 11217-11228.
- Xu, X., Ouyang, X.-k., & Yang, L.-Y. (2021). "Adsorption of Pb(II) from aqueous solutions using crosslinked carboxylated chitosan/carboxylated nanocellulose hydrogel beads." *Journal of Molecular Liquids*, 322: 114523.
- Yagub, M. T., Sen, T. K., Afroze, S., & Ang, H. M. (2014). "Dye and its removal from aqueous solution by adsorption: a review." *Advances in Colloid and Interface Science*, 209: 172-184.
- Yang, J., Ma, C., Tao, J., Li, J., Du, K., Wei, Z., . . . Ma, M. (2020). "Optimization of polyvinylamine -modified nanocellulose for chlorpyrifos adsorption by central composite design." *Carbohydrate Polymers*, 245: 116542.
- Yingkamhaeng, N., Intapan, I., & Sukyai, P. (2018). "Fabrication and Characterisation of Functionalised Superparamagnetic Bacterial Nanocellulose Using Ultrasonic-Assisted In Situ Synthesis." *Fibers and Polymers*, 19(3): 489-497.
- Yu, H., Zhang, S., Wang, Y., Yin, D., & Huang, J. (2021). "Covalent modification of Nanocellulose (NCC) by functionalized Graphene oxide (GO) and the study of adsorption mechanism." *Composite Interfaces*, 28(2): 145-158.
- Zarei, S., Niad, M., & Raanaei, H. (2018). "The removal of mercury ion pollution by using Fe₃O₄-nanocellulose: Synthesis, characterizations and DFT studies." *Journal of Hazardous Materials*, 344: 258-273.
- Zhan, C., Sharma, P. R., He, H., Sharma, S. K., McCauley-Pearl, A., Wang, R., & Hsiao, B. S. (2020). "Rice husk based nanocellulose scaffolds for highly efficient removal of heavy metal ions from contaminated water." *Environmental Science: Water Research & Technology*, 6(11): 3080-3090.
- Zhang, C., Jiang, Q., Liu, A., Wu, K., Yang, Y., Lu, J., . . . Wang, H. (2020). "The bead-like Li₃V₂(PO₄)₃/NC nanofibers based on the nanocellulose from waste reed for long-life Li-ion batteries." *Carbohydrate Polymers*, 237: 116134.
- Zhang, Z., Liu, X., Wang, H., He, H., & Bai, R. (2021). "Preparation and characterization of Enteromorpha prolifera nanocellulose/polyvinyl alcohol composite films." *Polymer Composites*, 42(4): 1712-1726.
- Zhong, C., Zajki-Zechmeister, K., & Nidetzky, B. (2021). "Reducing end thiol-modified nanocellulose: Bottom-up enzymatic synthesis and use for templated assembly of silver nanoparticles into biocidal composite material." *Carbohydrate Polymers*, 260: 117772.
- Zhou, L., & Xu, Z. (2020). "Ultralight, highly compressible, hydrophobic and anisotropic lamellar carbon aerogels from graphene/polyvinyl alcohol/cellulose nanofiber aerogel as oil removing absorbents." *Journal of Hazardous Materials*, 388: 121804.
- Zhou, X., Fu, Q., Liu, H., Gu, H., & Guo, Z. (2021). "Solvent-free nanoalumina loaded nanocellulose aerogel for efficient oil and organic solvent adsorption." *Journal of Colloid and Interface Science*, 581: 299-306.
- Zinge, C., & Kandasubramanian, B. (2020). "Nanocellulose based biodegradable polymers." *European Polymer Journal*, 133.