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Oxygen permeability properties of nanocellulose reinforced biopolymer nanocomposites

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ABSTRACT

Dry powder products are highly sensitive to oxygen and water vapour and must have a long shelf life. Currently, they are vacuum-packaged in sachets whose walls are made of a composite film that stops oxygen and water vapour from entering the package. Most of these composites are not biodegradable, a situation that the most agricultural company now wants to change to gain in sustainability. According to National Geography (2019), every year, about 8 million tons of plastic waste escapes into the oceans from coastal nations, with some estimates ranging to at least 400 years to break down. Over the years, there is huge interest in the development and use of biobased packaging materials, however, most of these materials are brittle and have low mechanical properties. Thus, in order to overcome these drawbacks, blended polymers with the reinforcement of nanocellulose are recommended. The aim of this paper is to review packaging films with oxygen barrier properties similar to those of the conventional films but also having biodegradable properties. From the literature, the oxygen permeability coefficient improved with the high strength, modulus, and ductility, as the nanocellulose was reinforcing with biopolymer. This might be due to nanocellulose that contributes to the higher stretchability of the composite biofilms. A fine dispersion of nanocellulose throughout the polymer host promoted several improved properties of the composite biofilms. In addition, nanocellulose had greatly enhanced oxygen and water vapour barrier properties. It would indeed push the usability of biopolymers forward, and certainly, prompt wider application of biodegradable polymers in the fields of environmental protection such as biodegradable food packaging.

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1. Introduction to oxygen barrier composite materials

Over the past few years, the replacement of petroleum-derived polymers with biopolymers has been one of the most important

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trends in the polymer field [1–4]. Among the drivers towards more sustainable packaging materials, the following ones stand out: the accumulation of plastics in the ocean, waste legislation, waste management and landfilling, marketing trends, growing environmental awareness, resource insufficiency, the need to reduce energy consumption, as well as producer and consumer accountability [5,6]. Biopolymers are recommended due to their biodegradability, availability, sustainability, biocompatibility, relatively low cost, non-toxicity and appealing physical and mechanical properties; in addition, they bring about a possible reduction in overall carbon footprint [7,8]. The utilisation of biopolymers as alternatives to petroleum-based plastics may reduce carbon dioxide emissions between 30% and 70% [9–11]. While the packaging industry is focusing on reducing the utilisation of raw material, waste volume, and transportation cost [12], producers are focusing on increasing shelf-life of the products and creating lighter products [13], and consumers are focusing on environmentally-friendly, green packaging, recyclable, and non-fossil-based packaging solutions [14]. The basic goal of a package is to protect the product in a sustainable way from the surrounding environment. Packaging materials function to provide mechanical, biological, and chemical protection to the product. Suitable packaging should make the product more sustainable, reduce the loss of product, enable long shelf life of the product, make the product safe, and fulfil performance metrics. Therefore, in order to achieve these requirements, barrier materials should protect against oxygen, grease, micro-organisms, water, aromatic compounds, moisture, and carbon dioxide. According to Helanto et al. [5], the most common challenges with biopolymers are their low resistance to gases, water, heat, and mechanical stress, as well as their relatively high price [15]. Compared to the common petroleum-based polymers, most biopolymers have some drawbacks, including low oxygen and water barrier properties as well as brittle structure due to the strong intermolecular and intramolecular hydrogen bondings between the chains, especially thermoplastic starch [16–21]. As a component of packaging materials, biopolymers such as polybutylene adipate terephthalate (PBAT) provides the necessary elasticity properties. However, it needs to be combined with other materials such as polylactic acid (PLA) to promote the required oxygen barrier performance [22]. Besides that, a commonly used approach to overcome these drawbacks is to blend these polymers with others polymers, additives or reinforcing agents (such as emulsifiers, kaolin, pectin, natural fibre, nanocellulose, bark, and others) to form composite materials [23–29].

Nanocrystalline cellulose (NCC) is a type of nanocellulose material that resulted from the chemical treatment of cellulose usually sulfuric acid [30–32]. A diameter of between 2 and 100 nm have been reported for nanocrystalline cellulose (NCC) and nanofibrillated cellulose (NFC) and their length can vary between tens of nanometres to micrometres. NCCs are mostly extracted from plants and agricultural residues including softwood, sugar palm, bamboo, jute fibres, coconut husk fibres, pineapple leaf, rice straw, and potato peel. Delignification with sodium hydroxide and sodium chlorite will result in the removal of lignin, hemicellulose, and other impurities. Following that, chemical treatment through acid hydrolysis will be conducted to produce nano-size cellulose. The hydrolysis conditions, including acid/fibre ratio, temperature, hydrolysis time, acid concentration, and sonication time show strong effects on the yield, sulphur content (in the case of sulphuric acid), anionic sites, Zeta potential, width, length and aspect ratio of the produced NCC [31,33]. NCCs show high tensile strength (>1 GPa) and high modulus (about 40 GPa). Researches on NCCs have experienced explosive growth in recent years due to their high potential in a wide variety of advanced applications, derived from the high strength, high modulus, large aspect ratio/surface area, rich surface chemistry, optical properties, low density and bio-

based nature of the nanofibers. Due to the advantages, nanocelluloses have been widely used as reinforcement materials in a variety of polymers including TPS, polylactic acid (PLA), polybutylene adipate terephthalate (PBAT), polyhydroxyalkanoate (PHA), polybutylene succinate (PBS), chitosan, and pectin [34,35].

This paper aims to give an overview of the use of nanocellulose reinforced biopolymer composite for packaging purposes as well as to summarise the recent developments in various barrier films based on nanocellulose with a special focus on oxygen barrier properties.

2. Types of nanocellulose

Nanocellulose as shown in Fig. 1, can be categorised into three types: (i) nanocrystalline cellulose (NCC), also known as cellulose nanocrystal (CNC) and cellulose nanowhiskers (CNW), (ii) nanofibrillated cellulose (NFC), also referred to as cellulose nanofibrils (CNF), and (iii) bacterial nanocellulose (BNC), also referred to as bacterial cellulose (BC) [36,37]. The types of nanocellulose are then merely dependent on the methods of isolation. These extraction methods can be divided into three types; (i) chemical methods (alkaline/acid hydrolysis, TEMPO mediated-oxidation), (ii) mechanical treatment (homogenisation, crushing, and machining) and (iii) combination of chemical and mechanical methods (electrospinning, hydrolysis). Whereas, BNC is derived explicitly via enzymatic or acid hydrolysis [38,39]. All nanocellulose divisions with their methods of isolation and properties are listed in Table 1.

3. Barrier properties of polymer

Permeability is the permeance multiplied by the thickness as shown in Table 2. Barrier properties are not only determined by the nature of the material but are also a function of temperature, pressure, and relative humidity. Barrier properties are usually measured under equilibrium moisture conditions with a controlled environment. According to Wang et al. [45], classification of commonly utilised polymeric packaging materials can be categorised into five categories, as shown in Table 3, which intentionally classifies polystyrene (PS), polyethylene (PE), and polypropylene (PP), into the poor oxygen barrier category.

4. Oxygen barrier properties of nanocellulose reinforced biopolymer

Recent progress has shown that nanocelluloses are able to serve as oxygen barrier materials (Table 4). Their unique characteristics have enabled them to form a dense microstructure, presenting high potential as a gas barrier. Coating a thin layer of TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl)-oxidised nanocellulose on films of PLA has resulted in the nanocomposite films with ultra-high transparency and outstanding oxygen barrier properties [53]. Besides that, dispersing nanocellulose in biopolymers has been shown to improve the oxygen permeability resistance [54–56].

The nanocellulose functions as a physical barrier structure, based on the “tortuous theory,” in order to prevent the movement of oxygen gas molecules through the barrier. According to Fukuzumi et al. [57], the oxygen molecules demonstrate kinetic widths of the same order of magnitude as the pores are partially separated in the nanocellulose network, in which they restrict oxygen from being transported through the films. This tortuous pathway varies according to the molecular diffusion and permeation features, free volume shape, quantity, and size. However, there is a possibility of existing free volume, in which this free volume is produced by the surface incompatibility between two components or is restricted

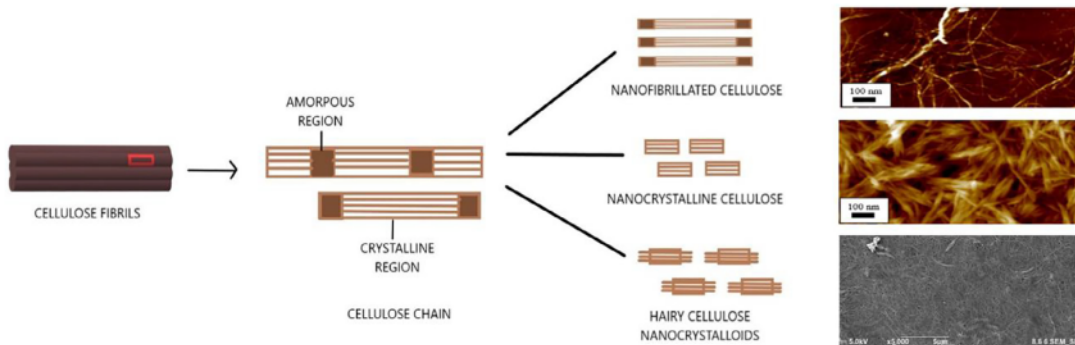


Fig. 1. Extraction of nanocellulose from cellulose fibre [14,31,39–42].

Table 1
Family of nanocellulose materials [38,43,44].

Type of nanocellulose	Synonyms	Typical sources	Extraction process	Formation	Average size	Properties
Nano/microfibrillated cellulose (NFC)	Microfibrillated cellulose, nanofibrils, microfibrils, nanofibrillated cellulose	Wood, high plants (sugar beet, potato) tuber, hemp, flax, sugar palm fibre	Mechanical processes	It can be extracted from cellulose chains using mechanical process to cleavage the fibre into nanometer size in diameter	D = 5–60 nm L = several mm Network structured nano-scaled fibres	Low crystallinity, longer length, high aspect ratio
Nanocrystalline cellulose (NCC)	Cellulose nanocrystals, crystallites, whiskers, rodlike cellulose, microcrystals	Sugar palm fibre, bamboo, kenaf, cotton, tunicin, wood, hemp, flax, wheat straw, ramie, Avicel, cellulose from algae and bacteria	Acid hydrolysis	It can be extracted from cellulose chains using acid hydrolysed amorphous region and left only crystalline region.	D = 4–70nm L = 90–450 nm Discrete rod-shaped particles	High crystallinity, short length and low aspect ratio
Bacterial nanocellulose (BC)	Bacterial cellulose, microbial cellulose, bio-cellulose	Low molecular weight sugars and alcohols, gram-negative bacteria	Bacterial synthesis	Different types of nanofiber networks	D = 20 – 100 nm	High aspect ratio

Table 2
Barrier parameters, equations, and units.

Barrier Property	Equation	Unit
Water Vapour Transmission Rate (WVTR)	$WVTR = \frac{\text{weight passed through}}{\text{area} \cdot \text{time}}$	$\text{g}/\text{m}^2 \cdot \text{day}$
Water Vapor Permeability (WVP)	$WVP = \frac{WVTR \cdot \text{thickness}}{\text{saturated pressure} \cdot \Delta RH}$	$\text{g} \cdot \mu\text{m}/\text{m}^2 \cdot \text{day} \cdot \text{kPa}$
Oxygen Transmission Rate (OTR)	$OTR = \frac{\text{volume gas used through}}{\text{area} \cdot \text{time}}$	$\text{cm}^3/\text{m}^2 \cdot \text{day}$
Oxygen Permeability (OP)	$OP = \frac{OTR \cdot \text{thickness}}{\text{oxygen partial pressure difference}}$	$\text{cm}^3 \cdot \mu\text{m}/\text{m}^2 \cdot \text{day} \cdot \text{atm}$

to the structures. Therefore, developing new high-gas barrier materials is still a challenge.

In addition to protecting the product against migration of the components and transmitted moisture, gas, and other dangerous agents from surroundings, an ideal barrier packaging should be thermally stable and mechanically robust [58,59]. The appropriate mixture of materials is crucial to improve the barrier properties of the packaging. The material's barrier properties as a function of the morphology of each component are displayed in Fig. 2 [60]. Lange et al. [60] summarised that the continuous layers arrange with multilayer structure (Fig. 2A) is more effective compared to the lamellar blend or fibrillar morphology (Fig. 2B); the

latter is better than the particulate system (Fig. 2C). It may be understood that mixtures include gaps between barrier particles to produce gas passageways to penetrate them, although the increased tortuosity reduces the penetration of gas. It can be concluded that although the increased tortuosity within the blends composite slows gas permeation, however, the formation of the gaps between the barrier particles would form passages for gases to penetrate through them. Therefore, this is the reason why many food barrier packaging industries utilise multilayer films with thin layers of continuous phase barrier materials. Besides that, literature data reviewed also revealed that the continuous nanocellulose phase in the neat and coated films performs better compared to the

Table 3
Barrier classifications of films based on oxygen and water vapour permeability^a.

Grade	Oxygen Permeability (cm ³ ·μm/m ² ·day·atm)	Example ^b	WVP (g·μm/m ² ·day·kPa)	Example ^b
Poor	>40000	HDPE 43307, PP 59055, PS 170866, PHB 302555 ^c , LDPE 790000 ^d	>3000	Silicone Elastomer 3502
Low	4000–40000	PVC 4252, BOPP 113204, PHA 150003, PLA 305,005	1000–3000	PA 6 1253, PLA 11,602
Medium	400–4000	EVOH wet 787, OPET 1181, PA 6 wet 1972, PET3543	40–1000	PS660, PHA 824 ^g PLA 898 ^f
High	40–400	PVdC 98, PA 6 dry 449	40–400	PAN 251, PVC 132, PET 79, LDPE 164 ^h
Very high	<40	EVOH dry 3.93	<40	HDPE 23, PP20, PVdC 17, OPET 4 ⁱ , BOPP 7 ^e

^aThe number by the name is the permeability coefficient of that material at 23–25 °C for OP without specified RH%; 37.8 °C and 90%RH for WVP. BOPP: biaxially oriented polypropylene; EVOH: ethylene vinyl alcohol; HDPE: high-density polyethylene; LDPE: low-density polyethylene; OPET: oriented polyethylene terephthalate; PA 6: polyamide 6; PET: polyethylene terephthalate; PHA: polyhydroxyalkanoates; PHB: polyhydroxybutyrate; PLA: polylactic acid; PP: polypropylene; PVC: polyvinyl chloride; PVdC: polyvinylidene chloride; PS: polystyrene; PAN: polyacrylonitrile. ^bRef [45]. ^cRef [46]. ^dRef [47]. ^eRef [48]. ^fRef [49]. ^gRef [50]. ^hRef [51] at 25 °C, 84/22%. ⁱRef [52].

Table 4
Oxygen permeability of nanocellulose reinforced biopolymer nanocomposites.

Formulation	Oxygen permeability	Conditions	Ref.
100% Bacteria CNCs/80% BCNCs + 20% PEG 900	6.1/3.6	24 °C, 0%	[61]
50% CMFs + 50% PVOH/25 CMFs + 25% PVOH + 50% clay	52264/62769	24 °C, 80%	[62]
	0.5/0.5	23 °C, 0%	
70% CMFs + 30% Sorbitol	6790/190	23 °C, 0%	[63]
	5200	23 °C, 80%	
75%PLA + 25%PHB/71% PLA + 24% PHB + 5% CNCs	13300/15300	RT, 0%	[64]
	0.6	23 °C, 0%	[65]
Bio-HDPE/CNF/bio-LDPE film	8	23 °C, 50%	[66]
	490	23 °C, 80%	
	242,219,981,208,338	23 °C, 50%	
		23 °C, 50%	
		23 °C, 50%	
Birch Xylan: Carboxymethylated CNFs: Plasticiser (G: Glycerol, M: MPEG, S: sorbitol)	70/30/0	23 °C, 50%	[66]
	60/40/0	23 °C, 50%	
	50/50/0	23 °C, 50%	
G: 35/35/30 M: 35/35/30 S: 35/35/30		23 °C, 50%	[66]
		23 °C, 50%	
PLA/PLA + 15%CNCS	25388/37644	23 °C, 0%	[67]
PLA-PBAT blend (modified) + nanocellulose	330	23 °C, 55%	[68]
Poly(3-hydroxybutyrate-co-3-hydroxyvalerate), PHBV	218.4	23 °C, 0%	[69]
Poly(vinyl alcohol-co-ethylene), EVOH	65.1	23 °C, 65%	[70]
Polycaprolactone (PCL)/PCL + 5%CNCS	8750/7350	23 °C, 0%	[71]
Poly(lactic acid (PLA)/PLA + 5% CNCS	30,500/17400	25 °C, 0%	[49]
Poly(lactic acid, PLA	541	23 °C, 55%	[68]
PVOH/CMFs: PVOH, 1:100	45000/50000	23 °C, 90%	[72]

^aOxygen permeability in cm³·μm/m²·day·atm, PEG: polyethylene glycol, PHB: polyhydroxybutyrate, PHOV: poly(vinyl alcohol), C: nanocellulose; CNFs: cellulose nanofibers; LDPE: low-density polyethylene; PBAT: polybutylene adipate terephthalate. Slash symbol “/” separates two formulations: the base material and nanocellulose composites. In the columns of oxygen permeability, the number before “/” is the oxygen permeability of the base material, and the number after “/” is the oxygen permeability of the nanocellulose composite.

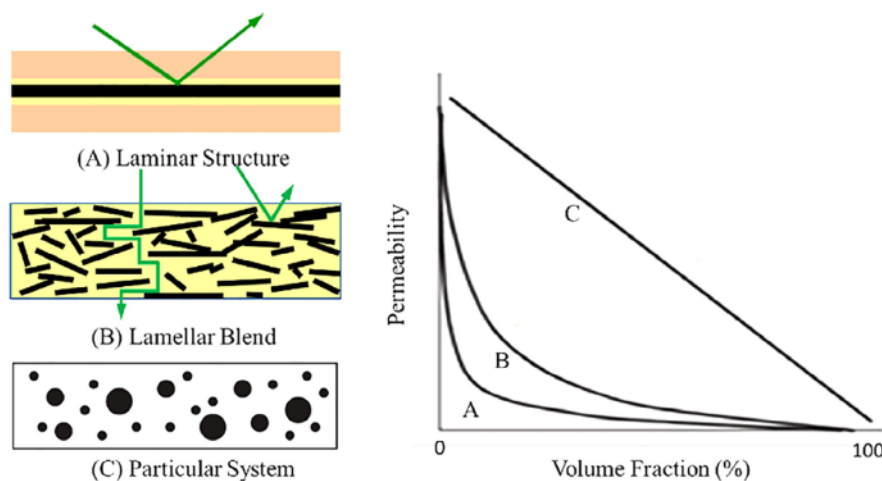


Fig. 2. Permeability of a blend of a high-barrier material as indicated in black in a low-barrier matrix as a function of the morphology [60].

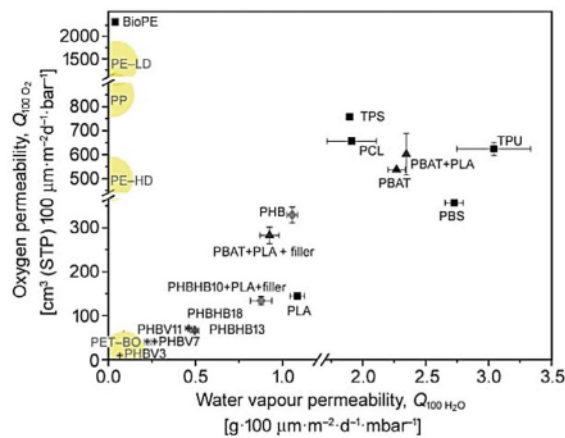


Fig. 3. Oxygen permeability $Q_{100\ O_2}$ versus water vapour permeability $Q_{100\ H_2O}$ for biopolymers with PHBV (●), PHBHB (◆), PBAT-based materials (▲) compared with conventional polymers (●): $Q_{100\ O_2}$ [cm^3 (STP) $100\ \mu\text{m}\cdot\text{m}^{-2}\cdot\text{d}^{-1}\cdot\text{bar}^{-1}$]/ $Q_{100\ H_2O}$ [$\text{g}\cdot 100\ \mu\text{m}\cdot\text{m}^{-2}\cdot\text{d}^{-1}\cdot\text{mbar}^{-1}$] of PE-LD (1.400/0.04), PE-HD (500/0.01), PET-BO (13/0.09), PP (850/0.02). Reproduced with copyright permission from Jost et al [73].

dispersed nanocellulose phase in nanocomposites with respect to gas barriers.

Besides that, based on Fig. 3, it can be observed that the reinforcement of the filler within the blended biopolymer improved the oxygen and water vapour permeability, and the result was much better compared to the commercialised petroleum-based polymer. The oxygen permeability coefficient of the PLA/PBAT/nanocellulose was expected to be as low as $100\ \text{cm}^3$ (STP) $100\ \mu\text{m}\cdot\text{m}^{-2}\cdot\text{d}^{-1}\cdot\text{bar}^{-1}$, combining with the high strength (30 MPa), Young's modulus (2 GPa), and elongation (400%), as indicated in Fig. 3, respectively. The suggested mechanism was the increased tortuosity resulting from the high crystallinity of nanocellulose.

5. Conclusion

The interest in nanocellulose reinforced biopolymer-based nanocomposite in packaging applications has accelerated significantly despite it is still at the infancy stage. A number of interesting solutions have been discussed or are being implemented at various stages such as types of nanocellulose and biopolymer used, and using nanocellulose as (I) laminar, (II) lamellar blend, or (III) in a particular system. The reinforcement of the nanocellulose with biopolymer-based packaging films strongly influence the oxygen barrier properties. However, an important necessity for further research is still needed due to inadequate resistance to moisture and water vapour. In addition, it is evident that there are many different combinations of contrasts between nanocrystal cellulose products, nanofibrillated cellulose products, and bacterial nanocellulose material. Such challenges and options implied that there will be a strong need for continuing research in the coming years in the attempt to achieve the full potential of nanocellulose in a wide range of packaging applications.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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