

## Chapter 27

# Nanocellulose-Polymer Composites: Novel Materials for Food Packaging Applications



A. K. Bharimalla, P. G. Patil, S. Mukherjee, V. Yadav, and V. Prasad

**Abstract** Nanocellulose is a revolutionary bio-based nanomaterial that possesses remarkable properties and has potential application in different industries. As a biodegradable filler in the manufacture of composite materials, coating and self-standing thin films, it offers novel and promising properties. There are fewer revisions focused on the use of nanocellulose-impregnated composite materials for different food packaging applications. Researchers have reported that the use of nanocellulose as a reinforcement in biopolymers and synthetic polymers improves the mechanical and barrier properties of the composite material. In this chapter we provide an exhaustive review of recent advances in the synthesis of nanocellulose and its application as a filler to produce nanocomposites for food packaging.

**Keywords** Biodegradable composite · Green composite · Nanocellulose biopolymer

## 27.1 Introduction

Packaging is an essential means to preserve the food quality and safety from manufacturing to its final use by the consumer. Food packaging helps reduce food waste during handling, transportation and storage by protecting food from physical, chemical or biological damage. Packaging is important for both consumers and marketers, as it not only provides different information such as ingredients, product characteristics, nutritional content, storage, but also helps improve customer acceptability and increase the product sale. Therefore, packaging is now considered the most important marketing tool in the modern economy. Around \$839 billion was the total turnover of the packaging industry in terms of revenue generation in 2015 (Anonymous n.d.), which is projected to increase at a rate of 3.5% on an average annually between

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2015–2020, reaching a value of \$998 billion in 2020 (Butschli 2016). The packaging sector is one of the fastest-growing sectors in India. India's share in global packaging is about 4%, which is equivalent to \$32 billion per year in 2015 and is expected to reach \$73 billion by 2020, with an annual growth rate of 18% (Anonymous n.d.). Factors such as the success in online marketing, the growth of organized retail, the increase in the consumption of ready-to-eat foods, the growth in flexible packaging market, the adoption of innovative packaging process and the development of machinery have contributed to the accelerated growth in the packaging industry. Of the total packaging sector, foods and beverages occupy the majority, accounting for 85%, followed by the plastic packaging market, which is expanding rapidly with a growth of 20–25% *per year* and is valued at 6.8 million of tons. The paper packaging industry reaches 7.6 million tons, and 10% is occupied by pharmaceutical products. Worldwide, among all packaging, food packaging has the highest share, i.e. \$161 billion, followed by other packaging such as beverage \$76 billion, pharmaceutical \$21 billion, cosmetic \$13.3 billion and another \$153 billion (Alam 2013).

The food industry has used petroleum-based plastic materials such as polyethylene as the preferred packaging material since last few decades (Gutiérrez and Alvarez 2017a). These materials were perceived as flexible, cheap, safe and versatile (Tice 2003), but they possess disposal limitations with very little recycling. In addition, rising petroleum costs, environmental concerns for disposal, globalization of food supply combined with consumer preference for fresher, convenient and safer foods have led to the emergence of novel sustainable packaging materials. These materials are an eco-friendly alternative to synthetic ones (Gutiérrez and Alvarez 2017b, c). Continuous research is being conducted to meet the consumer demands for new and innovative food packaging materials and technologies to produce sustainable packaging materials (Merino et al. 2018a, b; 2019a, b). These materials have changed the consumer's perception about raw materials, their selection, processing and consumption for different end-use applications. This is due to direct and indirect links of raw materials with general sustainability, energy efficiency and especially, security of supply. There is substantial growth potential for sustainable packaging materials for food packaging such as bio-based packaging products, biopolymers, bioplastics.

Bio-based food packaging materials are derived from renewable agricultural or marine sources (Álvarez et al. 2017, 2018). In the current era, green composite with bio-based material has gained preference as compared to petroleum-based composite due to the advantage of being natural, ecological and renewable (Satyanarayan et al. 2009). It has two main benefits, as they allow for a healthier ecosystem as well as rural employment along with economic development of the agricultural community. The limitation of biocomposites in the packaging industry is due to their poor mechanical and barrier properties (Azeredo 2009). In addition to this, their thermal properties are not at par with petroleum-based product. Further, processability is also one of the biggest limitations with current machinery due to low thermal stability and poor mechanical strength of biocomposites. These limitations can be addressed using advanced materials in nano-dimension (Gutiérrez et al. 2017a; Bracone et al. 2016; Gutiérrez et al. 2019; Toro-Márquez et al. 2018; Gutiérrez 2018a; Gutiérrez and Alvarez 2018, 2017d).

Nano-dimensional materials have some advanced properties due to their defect less structure and high strength properties. The intervention of nanotechnology not only solves the problems related to the properties, but also the cost economics (Sorrentino et al. 2007) and the problems of disposal of packaging materials. Nanocellulose which is a biopolymer, renewable and abundant, can be a twenty-first century solution as a filler material for bio-based composite material to be used for packaging industry (Mittal 2011). A detailed analysis of the nanocellulose production and its compatibility with hydrophobic matrix, as well as the application to produce nanocomposite for food packaging, was carried out in this chapter.

The water vapor permeability (WVP) of food packaging material is a critical issue in biopolymers, because they absorb water, which degrades the food quality. Therefore, the recent thinking process is addressing issues such as high WVP, poor mechanical strength, low thermal stability, improved physico-chemical and recyclability properties by using nanocomposite technology (Sorrentino et al. 2007). Materials with improved structure and properties can be processed by controlling important chemical and physical interactions of composites governed by surface properties (Gutiérrez et al. 2018a). The nanocomposites show different properties than their individual constituents, since a change in the diameter of the particles, layer thickness or fibrous material diameter to nanometer range influences the surface area-to-volume ratio (Ochsner et al. 2009). Different nanofillers have the potential to improve the migration of gases and the flavor properties of plastic packaging and increase the shelf life of the product, e.g. carbon nanotubes, kaolinite, graphene, nanoclays (Arora and Padua 2010)(Gutiérrez et al. 2017b).

Consumer demand and awareness about environmental problems due to plastic packaging has led to the development of novel bio-based and edible packaging materials (Gutiérrez et al. 2015a, b, c, d). Such materials will not only reduce packaging waste, but will also improve the quality of the stored food and, therefore, increase the shelf life of the product. Many thermoplastic polymers as starch can be used to prepare biodegradable packaging (Tharanathan 2003). Others include cellulose, chitosan (Cs)/chitin, protein (animal, plant based) or lipids (animal, plant derived) etc (Gutiérrez 2017a).

‘Active packaging’ (AP), a novel concept that has shown a paradigm shift in food packaging since the last two decades by shifting the protection function of packaging from passive to active. AP is a system in which the product, the package and the environment interact in a positive way to prolong shelf life of the food. AP actively changes the condition of the package to prolong shelf life or improve food safety or sensory properties, while maintaining the quality of the food. Active packaging is designed to provide chemical and physical benefits. The active packaging elements can be divided into different categories as absorber/scavenger, releaser, remover, time-temperature indicator and antimicrobial system. For any food, fresh vegetables and fruits, the absorption systems can be used as active packaging components to remove undesired gases and substances (oxygen, carbon dioxide, moisture, ethylene, flavor or odor, etc.) in order to extend the shelf life and prevent spoilage. The releasers consist of emitters of ethanol and carbon dioxide, preservative agents etc., removers help to eliminate lactose, cholesterol etc. of food, while temperature control systems can be self-heating or isolating materials.

Intelligent packaging (IP) is another innovative packaging system capable of carrying out intelligent functions such as detection, sensing, registration, tracing, communication and application of scientific logic, to improve food quality, extend the shelf life of the product, increase safety, provide information and warn about possible problems (Yam et al. 2005). IP system can for e.g. carry out intelligent functions as a release of an antimicrobial to protect the food product from spoilage. IP helps improve food safety and achieve the desired food quality. They can be tags or labels attached to a primary or secondary packaging as barcodes, RFID tags, time/temperature/gas indicators or biosensors (Gutiérrez et al. 2018b; Gutiérrez 2018b, 2017b; Gutiérrez et al. 2016a, b). A series of IP enabled technologies are available incorporating antimicrobial substance in nano-form to process polymer composites to control microbial surface contamination of foods. The successful production of viable edible films and coatings from whey proteins and their multi-functions as antioxidants, carrier of antimicrobials or other nutraceuticals with a significant primary barrier and mechanical properties add value for commercial applications in food industries (Ramos et al. 2012). In addition, due to the increasing demand in consumer for minimally processed products without preservatives, the IP technology has gained much attention from the food industry, as it uses low level of preservatives in packaging materials and ensures a minimal contact with the food (Cha and Chinnan 2004).

Many researchers have incorporated antimicrobial substances, sensors in nanocellulose-based polymer composites for packaging applications to detect and prevent food spoilage. Fortunati et al. (2012a) developed multi-functional bio-nanocomposite films of poly(lactic acid) (PLA), cellulose nanocrystals (CNCs) and silver nanoparticles. These films showed a constant antibacterial effect against *Staphylococcus aureus* and *Escherichia coli*, with potential use in food packaging applications. El-Wakil et al. (2015) developed wheat gluten/nanocellulose/titanium dioxide composite papers for active food packaging. These coated papers showed good antimicrobial activity against Gram positive and negative bacteria, and yeast.

## 27.2 History of Packaging Industry

The need to store food arose with the progress in human civilization when humans began to travel long distances in search of food. The oldest packaging materials used for this purpose were pumpkin's, shells and leaves. Subsequently, humans devised different innovative nature-derived packages with an increase in the need for storage and packaging purposes, e.g. baskets woven from leaves and barks, hollowed out vegetables and wooden logs, different animal parts such as skin/hide, organs etc. During 1500 BC–500 AD, new packages such as ceramic vessels and amphorae arose in the Mediterranean region for the commercial transport of wine and other products. The different advances in food packaging over the years helped to protect food from external contact such as contamination by microbes, thus reducing its growth, in addition to its basic function as a means of consumption and storage. The additional packaging materials developed over the years prevent

contact of food with change in air, pH and moisture, which can lead to microbial degradation of food (Raheem 2012).

Glassmaking started as a type of ceramics in 7000 BC and the glassmaking technique was industrialized around 1500 BC in Egypt. Around 7000 BC, colored water pots were used as containers to store food and water. By 1200 BC, the cups and bowls were made by pressing the glass into molds. Transparent glass was invented in the Christian era and its manufacturing process spread across the European continent for the next 1000 years. The technique was refined even more during eighteenth and nineteenth centuries. The first automatic machine to manufacture rotary bottles was invented and patented by Owens in the year 1889. During 1900s–1960, liquid products were stored mainly in glass containers.

The oldest form of flexible packaging was paper. The paper was invented in China during 200 BC–220 AD. The word paper is derived from the Greek word Papyrus. The paper-based packaging was first used by the Chinese to wrap foods with the use of treated mulberry bark during 1st and 2nd century BC. The paper-making technique was further refined over the next 1500 years and the knowledge travelled westwards from China to Central Asia, the Middle East, and Europe (Welt 2005). Papermaking technique was introduced in the United Kingdom in 1310, while it came Pennsylvania, USA in 1690 (Berger 2002).

Paperboard was first used to manufacture folding cartons in the 1800s, first paperboard carton/cardboard box was made in England in 1817 and its thinner type paperboard carton was later used for cereal storage. The use of paper in packaging began with the development of paper bags. The commercial production of paper bags began in Bristol, England in 1844. The corrugated boxes used widely today as shipping container to hold a series of smaller packages were developed in the 1850s (Risch 2009). In 1852, the machine to manufacture paper bags was invented and patented by Francis Wolle in United States of America (USA), which led to development of glued paper sacks and gusset design in 1870s, which are also used today. In 1870s, the world's first semi-flexible packaging was accidentally developed by Robert Gair, who invented the first automatic-made cardboard. These folding cartons used to date form the backbone of dry and processed food products. In 1905, machines were invented to produce in-line printed paper bags.

Initial metal-based packaging materials as boxes, cups and containers made from gold and silver were precious/too expensive for ordinary people to use in the ancient era. Other metals emerged progressively as common packaging materials with discovery of cheaper metals for the processing of strong alloys, coatings and thinner gauges and large-scale production (Hook and Heimlich 2011). In 1805, Nicholas Appert (“Father of Canning”) proved that food could be sterilized and preserved for an extended time by boiling at high temperatures and then sealing in tin containers. Later, in the year 1810, inventor and merchant Peter Durand received a patent for developing a sealed cylindrical can made from tinplate for food storage. The first beverage can made of tin-plated steel was made by Kruger Beer in 1935, until then the drinks were available in glass bottles. In 1959, Coors introduced the aluminium can (which is currently used in carbonated beverages) and in the same year, Emral Frazee invented the pop tab. The first ring pull was introduced in 1963, which facilitated the opening of the can and provided ease of drinking

directly from it. The stay tab which is a ring tab that stays attached to the can, was introduced in 1975.

Throughout the 20th century, paper and paperboard packaging increased in popularity and, subsequently, in the late 1970s and 1980s with the arrival of plastics as an important element in packaging, paper and paper-based products were replaced. The use of plastics in packaging began in the 1950s. The packaging industry is the largest user of plastics because more than 90% of flexible packaging is made of plastics and only 17% of rigid packaging. Generally, barrier resins are used for the preparation of plastic containers to improve product protection and make them more cost effective. Plastic materials made up of large organic molecules are useful products, since they are fluid, can be sealed with heat, are easy to print and can be integrated into production processes (Marsh and Bugusu 2007). The application of plastics in packaging has increased worldwide with an estimate of 280 metric tons (Paine and Paine 2012).

The molded deodorizing squeeze bottles were introduced in 1947 and in 1958 for packaging; while heat shrinkable films were developed from the mixture of styrene with synthetic rubber. Cellulose acetate first derived from wood pulp in 1900 and developed for photographic uses in 1909. New manufacturing protocols were also developed using various techniques such as forming, molding, casting and extrusion to prepare plastic products in large quantities (Plastic Make It Possible Report 2010).

In 1977, after the introduction of polyethylene terephthalate (PET) containers, it became easy to manufacture beverage containers, which are mainly used today. Other novel materials, such as transparent films and cellophanes, have been used as external wrappers to maintain the shape. By 1980, foods and other hot-fill products, such as jams, could also be packaged in PET. In 1986, aluminium trays were replaced by plastic microwavable trays. In 1996, metallocene-catalyzed polyolefin was introduced to reduce food waste. In 2000, the entry of PLA into packaging market signalled the return of biobased plastic (Vink et al. 2004).

Active packaging is a new area of research which involves the combination of food packaging materials with antimicrobial substances. With the advent of nanotechnology, it is much easier to functionalize polymer films based on nanoparticles with antimicrobial agents for food packaging. The nanocomposites are much more superior compared to conventional packaging materials in terms of barrier properties, mechanical strength and heat resistance (Herniou--Julien et al. 2019). The polymer nanocomposites for food packaging have been considered for the entire life cycle of the packaging material (Silvestre et al. 2011). The life cycle is a very important property in the packaging material from procurement, processing, transportation, delivery and disposal of raw materials (Chaffee and Yoros 2007).

### 27.3 Polymer Matrix

Fiber along with a suitable polymer matrix, provide good strength and adhesion to a composite material (Deo 2010). Generally, the matrix is deformed in the load application or load transfer. Therefore, when the matrix is reinforced with any

fibrous filler, the applied load is transferred mostly to the fiber and provides an adequate load bearing capacity. The matrix redistributes the load to the surrounding fibers and restricts the buckling of the fibers due to compression (Outwater 2014). Polymer matrices are broadly divided into two categories: thermoset and thermoplastics.

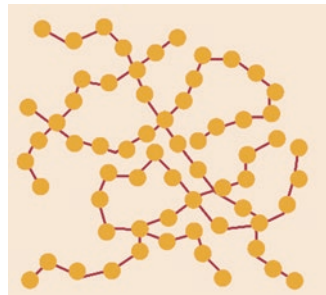
### 27.3.1 Thermoplastic Polymer Matrix

Plastics that melt upon being heated and solidify upon cooling are known as thermoplastics which are generally solid at room temperature (Fig. 27.1). There is no chemical change in any number of melting and solidification of these plastics (Morena 1988). But a greater number of heating cycle degrades the polymer. Examples of thermoplastic polymers are unsaturated polyesters (UPs), polyetherimide, polyamide imide, polyphenylene sulfide, polyetherketone and liquid crystal polymers. Thermoplastics are mainly used in high volume industries, such as the automotive and aviation industries, etc. After adequate reinforcement with plastic, carbon or graphite fiber, they can be used instead of epoxy in the next generation aircrafts (Astrom 1997).

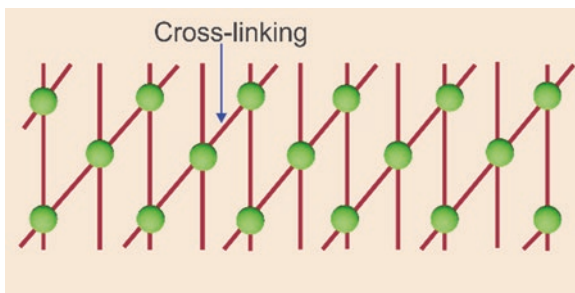
### 27.3.2 Thermoset Polymer Matrix

The polymeric materials after a chemical reaction are transformed from liquid to solid state, when the material is not cured, the materials are very small unlike molecules, i.e. monomers. After molecular cross-linking during the reaction, longer molecular chains are formed, leading to solidification (Fig. 27.2). These thermoset polymeric materials are permanent and irreversible. The different types of thermoset polymer matrices used in composites are bismaleimides, epoxy (epoxide), phenolic (PF), UP, polyimide, polyurethane (PU) and silicone (Astrom 1997).

**Fig. 27.1** Polymerized thermoplastic polymer



**Fig. 27.2** Polymerized thermoset polymer



## 27.4 Cellulose

In 1839, the French chemist A. Payen used the term “cellulose” for the first time in his report of the French Academy (Payen and Hebd 1838). The French chemist Anselme Payen in 1838 described a resistant fibrous solid, which remained after treating various plant tissues with acids and ammonia (Klemm 2005). The molecular formula ( $C_6H_{10}O_5$ ) was determined by elemental analysis and the term “cellulose” was used to refer it. It was used in the form of wood, cotton and other vegetable fibers as an energy source, for building materials and clothing for thousands years since its discovery (Doree 1947). Cellulose is a long-chain natural polymer made by linking of smaller molecules (Fig. 27.3). These links in the cellulose chain consist of sugar,  $\beta$ -D-glucose (Alemdar and Sain 2008). Cellulose is found in cotton and all lignoncellulosic biomasses. Lignoncellulosic agricultural by-products can be used to obtain cellulose, among them: the residues of wheat and cereals (Jahan et al. 2011), jute (Nelson et al. 2000), soybean husk (Bochek et al. 2003) flax fibers and flax straw (Bhattacharya et al. 2008), sugarcane bagasse (Istvan and Plackett 2010), corn, sorghum, barley, pineapple, sorghum, bananas, coconut crops, etc. (Hubbe et al. 2008). Despite its various use in the fiber, paper, films and polymer industries, the use of cellulose has generated interest in the processing of novel material applications due to its super functionalities, extremely large, active surface area and low cost (Turbak et al. 1983).

### 27.4.1 Nanocellulose

In the early 1980s, the term nanocellulose was publicly used for the first time by Klemm (Klemm et al. 2011). Turbak, Snyder and Sandberg in the late 1970s at the ITT Rayonier labs in Whippany, NJ, USA used the microfibrillated nanocellulose (MFC) terminology to describe a product prepared as a gel-like material. This material was prepared by passing wood pulp through a Gaulin-type milk homogenizer at high temperatures and high pressures impacting against a hard surface through the ejection process. Nanocellulose is a promising innovative material for the modern industry of the twenty first century. In future, synthetic fillers will be replaced by nanocellulose to reinforce polymer composites for use in the automotive industry, packaging and furniture production Jahan et al. (2011).



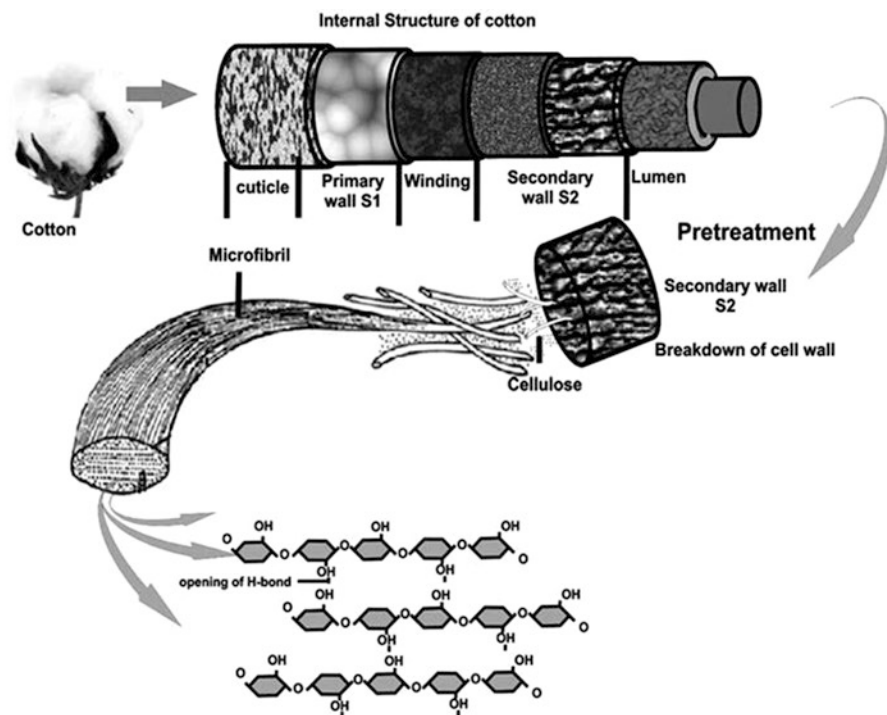



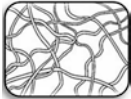
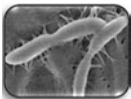
Fig. 27.3 Structural organization of the plant cell wall



Fig. 27.4 Image of nanocellulose prepared from wood pulp

The word 'nanocellulose' generally refers to cellulosic materials with a dimension in the nanometer range and look like gel if it is prepared from wood pulp as shown in Fig. 27.4. Depending upon the raw material, the production method and its size, nanocellulose is classified into three main categories (Iwamoto et al. 2009). The three main types of nanocellulose are cellulose nanocrystal (CNC), cellulose nanofibril (CNF) and bacterial nanocellulose (BNC). The nomenclature used by Klemm (2005) is given in Table 27.1.

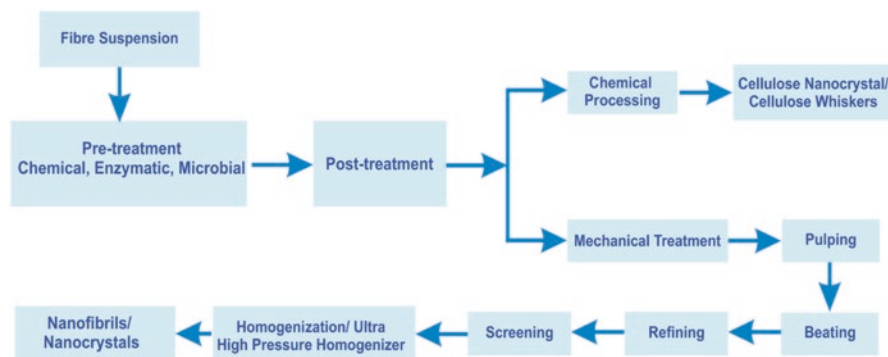
**Table 27.1** Classification of nanocellulose (Iwamoto et al. 2009; Gama et al. 2012; Cherian et al. 2010)

Type of nanocellulose	Synonyms	Typical sources	Average size	Images
Cellulose nanocrystal	Nanocrystalline cellulose, whiskers, rod like cellulose, microcrystals	Wood, cotton, hemp, flax, wheat straw, rice straw, mulberry bark, ramie, MCC, Avicel, tunicin, algae, bacteria, etc.	Diameter: 5–70 nm Length: 100–250 nm (from plant); 100 nm several micrometers (from cellulose of tunicates, algae, bacteria)	
Nanofibrillated cellulose	Nanofibrils, microfibrils, nanofibrillated cellulose, microfibrillated cellulose	Wood, sugar beet, potato, tuber, hemp, flax etc.	Diameter: 5–60 nm Length: several micrometers	
Bacterial nanocellulose	Microbial cellulose, biocellulose	Low-molecular-weight sugars and alcohols	Bacterial synthesis Diameter: 20–100 nm	

### 27.4.2 Preparation and Properties of Nanocellulose

The most general method for preparation of nanocellulose is top-down approach using physics, chemistry and chemo-mechanics. The detailed scheme of this method is shown in Fig. 27.5. BNC is being synthesized by a bottom-up method from the glucose by a family of bacteria (Iwamoto et al. 2009), yeast, algae, etc. In this chapter, we mainly focus our attention on nanocellulose produced by the top-down method from wood or agricultural/forest crops or residues, i.e. lignocellulosic biomass.

Several methods of preparing nanocellulose from cellulosic materials have been reported, such as steam explosion treatment (Mandal and Chakrabarty 2011), acid or alkaline hydrolysis (Moran et al. 2008; Henriksson et al. 2007), enzyme-assisted hydrolysis (Chen et al. 2011), as well as a combination of two or several of the aforementioned methods (Keeratiurai and Corredig 2009). High-pressure homogenization (HPH) due to its simplicity, high efficiency and no requirement of organic solvents is an efficient technology for biomass refining (Kaushik and Singh 2011). Typically MFC is produced as a suspension in water. The viscosity of the suspension changes during homogenization from a low-viscosity to a high. Normally, a 2% fiber suspended in water is used for the preparation of MFC. To minimize the size of the cellulose fibers and to avoid clogging during homogenization, pre-treatment of cellulose, such as steam explosion, microfluidization processor or other methods is essential (Lee et al. 2009a; Zimmermann et al. 2010; Brandt et al. 2010). Recently, ionic liquids at room temperature have emerged as new agents for pre-treatment, since they have an excellent dissolving capacity of cellulose (Li et al. 2012).



**Fig. 27.5** Flow diagram for the synthesis of nanofibrils and nanocrystals by mechanical and chemical routes

At higher concentrations, the increase in viscosity during processing becomes too high to continue processing. Therefore, a schematic overview of approaches to produce nanocellulosic materials in low concentration from fibers is provided in Fig. 27.5 (Siqueira et al. 2010; Lavoine et al. 2012).

Many intrinsic properties have been observed in MFC, such as low density, high chemical reactivity, high strength and modulus, and high transparency that make it attractive for applications (Nogi et al. 2009; Lee et al. 2009b; Paakko et al. 2007; Siro and Plackett 2010).

Recently, a great interest has been attracted to nanocellulose as a potential filler for use in nanocomposites. A wide range of polymer matrices can be incorporated with nanocellulose as waterborne polyurethane, poly(3-hydroxybutyrate), hydroxypropyl cellulose, poly(L-lactide), poly(3,4-ethylenedioxythiophene), polyvinyl acetate and poly(*o*-ethoxyaniline). Table 27.2 shows that the nanocellulose stiffness and tensile strength (TS) are comparable to those of aramid fiber (Kevlar) and better than glass fiber, which are used commercially to reinforce plastics.

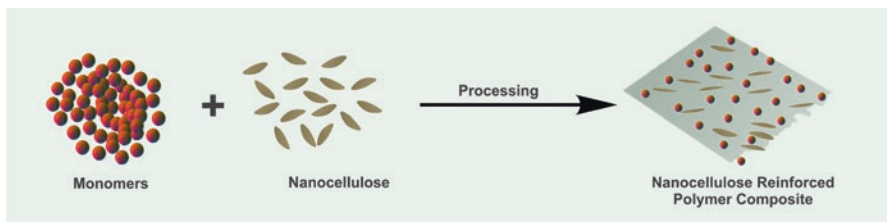
Nanocellulose films exhibit high strength (> 200 MPa), high stiffness (~20 GPa), and high strain (12%). The strength/weight ratio of nanocellulose is 8 times more than stainless steel (Aulin et al. 2010). Nanocellulose composites can thus be used as coatings and films, paints, foams and packaging materials (Cai et al. 2011).

## 27.5 Nanocellulose-Polymer Composite Films

Nanocomposite is now an expanding research area that offers new materials with novel functional properties (Ruiz et al. 2000). Nanocomposites are made up of nano-size fillers with particular size, dimension and surface chemistry properties. Nanocellulose is used mainly with hydrocolloids such as starches, poly-saccharides, proteins, pectins and synthetic polymers. Different inorganic nanoparticles have

**Table 27.2** Mechanical properties of various reinforcements

Sl. No.	Plant fibre/materials	Strength (GPa)	Modulus (GPa)	Reference
1	Cellulose micro/nanofibril	10	150	Sakurada et al. (1962)
2	CNC	10	150	Revol et al. (1998)
3	Aramid fibers (Kevlar)	3–3.5	130	Denoyelle (2011)
4	Cotton	0.3–0.7	6–10	Cristaldi et al. (2010)
5	Kapok	0.093	4	Cristaldi et al. (2010)
6	Bamboo	0.57	27	Cristaldi et al. (2010)
7	Flax	0.5–0.9	50–70	Cristaldi et al. (2010)
8	Hemp	0.31–0.75	30–60	Cristaldi et al. (2010)
9	Jute	0.2–0.45	20–55	Cristaldi et al. (2010)
10	Aluminum wire	0.62	73	Bledzki and Gassan (1999)
11	Steel	0.54	200	Bledzki and Gassan (1999)
12	Kenaf	0.29–1.19	22–60	Bledzki and Gassan (1999)
13	Ramie	0.91	23	Cristaldi et al. (2010)
14	Abaca	0.012	41	Cristaldi et al. (2010)
15	Banana	0.53–0.92	27–32	Cristaldi et al. (2010)
16	Pineapple	0.413–1.627	60–82	Cristaldi et al. (2010)
17	Sisal	0.08–0.84	9–22	Cristaldi et al. (2010)
18	Coir	0.106–0.175	6	Cristaldi et al. (2010)
19	Ramie	0.4–0.94	61.4–128	Cristaldi et al. (2010)
20	E-glass	2–3.5	70	Westman et al. (2010)
21	Carbon	4	230–240	Westman et al. (2010)

**Fig. 27.6** Preparation of polymer–nanocellulose composite

been recognized as possible additives to improve the polymer performance (John and Thomas 2008). A polymer composite is a combination of a polymer matrix and a strong reinforcing phase, or filler (Fig. 27.6). Polymer nanocomposites are polymers (thermoplastics, thermosets or elastomers) that have been reinforced with small amounts (less than 5% by weight) of nanosized particles having high aspect ratios ( $L/D > 300$ ) (Denault and Labrecque 2004).

### 27.5.1 Preparation of Nanocellulose-Based Composite Films

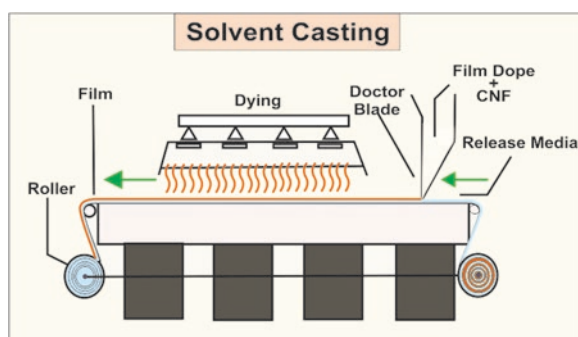
Polymers derived from petrochemical predominate in food packaging due to their easy processing, excellent barrier properties and low cost (García et al. 2004). The use of nanocellulose acting as an active substance carriers, such as antioxidants and antimicrobials helps to extend the food quality and safety, thus extending the shelf life of them. There is potential for the processing of nanocellulose-based polymer composites for preparing cheap and lightweight nanocomposites for food packaging applications. In the recent years, advanced research and development in the field of polymer science and the extensive use of advanced technology for the preparation of polymers have generated greater interest in the preparation and characterization of novel polymer materials and their composite films.

#### 27.5.1.1 Solvent Casting Method

One of the simplest methods used for the preparation of polymer nanocomposites is the solvent casting method, since it requires simple processing equipment. The nanocellulose reinforcement in the solution casting is dispersed within a given medium (0.05–5 wt% solids), water or different organic media to prepare a homogeneous nano-scale aqueous suspension by stirring at room temperature or using an autoclave reactor to mix at high temperatures and then polymer solutions are mixed with it. The composite films are subsequently produced with this mixture *via* casting on a suitable surface, followed by an evaporation/drying in oven under vacuum (Fig. 27.7) (Ghosh and Sain 2014).

This processing method strongly influences the mechanical properties of the resulting nanocomposite. It has been observed that solution casting of nanoscale reinforcements, especially with an aqueous latex dispersion matrix, produces superior results compared to melt-extruded composites due to better dispersion of nanoscale cellulose reinforcements and the possibility to form hydrogen bonds between reinforcements and matrix material (Favier et al. 1995). It is a low-temperature environmentally friendly process which requires a small amount of

Fig. 27.7 Solvent casting



sample to produce films of uniform thickness. However, the method has several limitations, since it consumes time, is useful when a very small amount of reinforcement is required, is limited to the laboratory scale and can involve complex processes with a high energy consumption (Khoskhava 2014).

The solution casting method is used to process polymer composites with nanocellulose reinforcement in water soluble polymer matrices such as poly(oxyethylene) (PEO) (Samir et al. 2004), polyvinyl alcohol (PVOH) (Roohani et al. 2008), water-borne PU (Guangjun et al. 2008), starch (Teixeira et al. 2009) and water-insoluble polymer matrices as polypropylene (De Menezes et al. 2009), polyvinyl chloride (Chazeau et al. 1999), polyvinyl acetate (Garcia de Rodriguez et al. 2006), polycaprolactone (Siqueira et al. 2009) and PLA (Sanchez-Garcia and Lagaron 2010; Yu et al. 2008; Lin et al. 2009).

### 27.5.1.2 Melt Intercalation Process

This is the most promising and practical method for the preparation of polymer nanocomposites. Vaia, Ishii and Giannelis (1993) first used the melt intercalation process to process polymer composites in 1993. Since solvents are not required, this method is used in the polymer processing industry using compounding devices such as extruders or mixers. Melt intercalation is a top-down method of polymer processing. This simple economic and environmentally friendly processing method involves direct mixing of nanoreinforcement with molten polymer to optimize the polymer–nanomaterial interactions. The nanocomposite is formed when the polymer–filler mixture is hardened above the glass transition temperature ( $T_g$ ) of the polymer (Fig. 27.8).

The polymer chains penetrate into the filler/reinforcement layers inducing intercalation. According to the compatibility between the surface of the layers and the polymer, either intercalated or exfoliated nanocomposites can be obtained (Kumar et al. 2009). This method results in normally lower levels of exfoliation

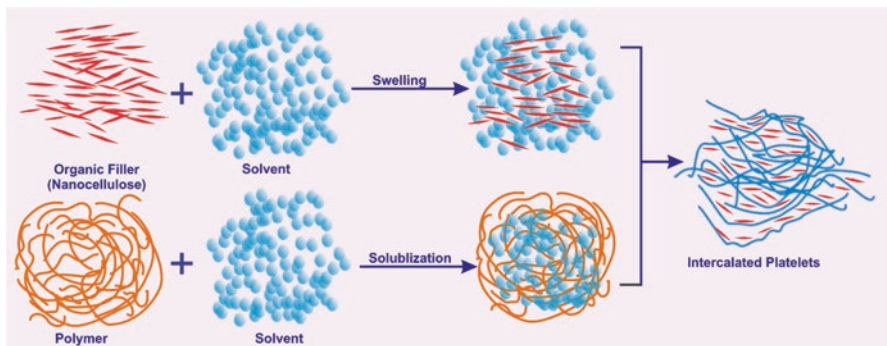


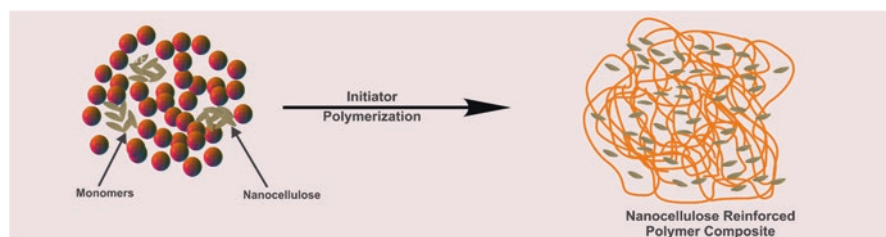
Fig. 27.8 Melt intercalation process for preparation of nanocomposite

compared to *in situ* polymerization. Polymers not suitable for solution intercalation, adsorption or *in situ* polymerization methods, can be used to prepare nanocomposites by this method (Hussain et al. 2006). The melting intercalation process of the polymer preparation reduces the interfacial tension and improves matrix-filler interactions. The method is more flexible and does not require chemical reaction or solvent (Liu et al. 1999; Karande 2013).

### 27.5.1.3 *In situ* Polymerization

The first method used to synthesize nanocomposites was *in situ* polymerization. The *in-situ* polymerization of monomers in the presence of cellulosic nanofillers is advantageous as compared to the traditional processing methods mentioned above, since a uniform dispersion of cellulosic nanofillers in the polymer matrix can be achieved by minimizing aggregation by their improved interaction with growing polymer molecules. This method also reduces the moisture absorption behavior of wrapped cellulosic nanofillers and improves the biodegradability of nanocomposites under compost conditions when required for specific applications such as packaging. However, this method is applicable only when the polymerization is carried out in liquid phase where liquid monomer molecules are polymerized in presence of nanocellulose filler. This process is conventionally used to synthesize thermoplastic nanocomposites, while for thermosets such as epoxies or UPs, a curing agent or peroxide is added to initiate the polymerization. Either the addition of curing agent or the increase in temperature can initiate polymerization for the thermosets (Fig. 27.9) (Messersmith and Giannelis 1995).

This processing method can be scaled up to an industrial level and can dramatically increase the industrial development of cellulosic nanocomposite products. Several researchers have used the *in situ* polymerization method to develop novel nanocellulose based composites with new potential applications, e.g. polymethylmethacrylate (PMMA) (Mabrouk et al. 2011; Maiti et al. 2013), polyacrylamide (PAAm) (Dufresne et al. 2003), PANI (Zhou et al. 2011), polyurethane (Lacerda et al. 2013), polypyrrole (Nystrom et al. 2010).



**Fig. 27.9** Process for the preparation of nanocomposite by *in situ* polymerization

### 27.5.1.4 Ring-Opening Polymerization

The ring-opening polymerization (ROP) reaction is of particular interest because it finds wide application in thermostable polymers. By means of this technique, epoxy resin networks or cross-linked UPs can be synthesized, which are of great importance in the market of thermoset polymer-based composites.

ROP is a well-known technique to polymerize cyclic monomers such as lactones and lactides, where alcohol generally acts as an initiator. This method is used for the polymeric modification of cellulose and various cellulosic derivatives (Jerome and Lecomte 2008). Depending on monomers, catalysts, initiators used, the polymer is prepared through different mechanisms using ROP method (Fig. 27.10). Several scientists have used ROP for the preparation of nanocellulose-based polymer composites (Goffin et al. 2011; Peltzer et al. 2014; Habibi and Vignon 2008; Chen et al. 2009; Lonnberg et al. 2011; Braun et al. 2012). The nanocomposite sheets produced by ROP possess good mechanical strength.

The ROP approach was applied for the first time to the CNCs by Habibi et al. (2008) who grafted polycaprolactone onto the surface of CNCs using stannous octoate ( $\text{Sn}(\text{Oct})_2$ ) as a grafting and polymerization agent. Chen et al. (2009) and Lin et al. (2009) conducted similar ROP under microwave irradiation to improve graft efficiency and incorporated PCL-CNCs into the PCL matrix. Lonnberg et al. (2011) grafted PCL chains on nanofibrillated celluloses (NFCs) and the resultant PCL-grafted NFC films were prepared by thermoforming in the form of bilayer laminates. Oksman et al. (2006) reported a direct correlation between the length of grafted PCL chains and interfacial toughness of the resulting laminate.

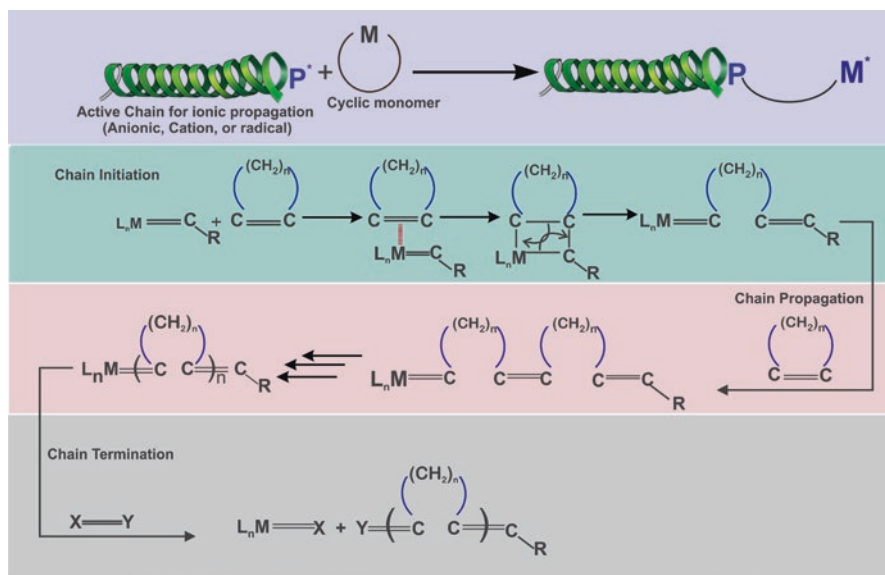


Fig. 27.10 Ring opening process for the preparation of nanocomposite



Extrusion methods are being used for the formation of nanofiber-reinforced polymer composites. Some reviews related to the preparation of composite films are highlighted below:

Oksman et al. (2006) reported on the preparation of PLA nanocomposites reinforced with cellulose whisker using melt extrusion method. In this method, the nanocellulose polymer composite was prepared by pumping nanocrystal suspension into the melt of the polymer during the extrusion process (Oksman et al. 2006). The authors observed that the hydrophilic nature of the cellulose results in the formation of additional hydrogen bonds between amorphous parts of the nanoparticles leading to an irreversible agglomeration during drying and aggregation in nonpolar matrices (Oksman et al. 2006). The film composites produced show a higher Tensile Strength (TS) as the proportion of modified cellulose increases and the elongation at break increases as the proportion of PLA increases. So modified cellulose plays a vital role in increasing the TS of composite films. These films can thus be used as a package to protect food from oxidation reaction and moisture (Sandeep et al. 2012).

Bruce et al. (2005) prepared composite materials using MFC from Swedish roots and different resins, including four types of acrylic and two types of epoxy resins. It was found that the stiffness and the strength of all the composites were significantly higher than the unmodified resins. Using this method, nanocomposites with good mechanical properties were prepared using vegetable pulp with a wide range of resins (Bruce et al. 2005). Low coefficient thermal expansion (CTE) is another beneficial feature of nanocellulose. It has been reported that the CTE values for nanocellulose are as low as 0.1 ppm/K, which is comparable to that of quartz glass. The low CTE for nanocellulose in combination with high strength and Young's Modulus (YM) makes it a potential reinforcement material for the manufacture of solar cells, actuators, flexible displays, electronic papers, panel sensors etc. (Nishino et al. 2004). Nogi and Yano (2008) prepared nanocomposites using transparent acrylic resin (with low YM) with 5% bacterial cellulose (BC). The ductile, foldable and transparent nanocomposite showed a low CTE and a high YM. A film stacking method was used to prepare PU-MFC composite materials, where the PU films and nonwoven cellulose fibril mats were stacked and compression molded (Seydibeyoglu and Oksman 2008). Wan et al. (2006) used PVOH reinforced with BC as a reinforcement material for medical device applications. The authors developed a PVOH-BC nanocomposite with mechanical properties over a wide range, thus making it appropriate to replace different tissues (Wan et al. 2006).

Khan et al. (2014) prepared methylcellulose (MC) films by using the casting methodology in a 1% aqueous solution containing 0.5% vegetable oil, 0.25% glycerol and 0.025% Tween-80. These authors studied the effect of gamma radiation on the nanocellulose-containing MC-based composites and observed that mechanical properties of the films increased slightly at low doses due to the reorientation of the nanocellulose fibers (NCFs), whereas barrier properties were improved even more. Dufresne et al. (2000) and Dufresne and Vignon (1998) prepared and reported that the biodegradability of modified potato thermoplastic starch (TPS) composite films containing MFC obtained by the solution casting methodology were preserved, and that MFC significantly reinforced the starch matrix, regardless of the plasticizer

content, and increase in YM depended on filler content almost in linear manner. It was also observed that YM and TS were significantly improved in the nanocomposite films due to the uniform dispersion of the nanofibers in the polymer matrix. Azeredo et al. (2010) developed composite films by reinforcing the nanocellulose in Cs and glycerol as a plasticizer. Pereda et al. (2010) developed sodium caseinate films with nanocellulose by dispersing the fibrils into film-forming solutions, casting and drying. The composite films were found to be less transparent and more hydrophilic than pure sodium caseinate films. The caseinate films showed an initial rise and subsequent decrease in WVP with increase in filler content.

## 27.6 Characterization of Composite Films

Different characteristics such as surface morphology, mechanical, thermal and optical properties, as well as water and gas permeability using various instrumental techniques are conducted to evaluate the properties of nanocomposites. Microscopy helps to study and understand the distribution of nanocellulose in the polymer matrix and interfacial adhesion.

### 27.6.1 Scanning Electron Microscopy (SEM)

The morphological and macroscopic study of nanocomposites is carried out by using scanning electron microscopy (SEM). An electron beam scan focused across the surface of the sample results in signals that are converted into an image on a computer screen. The most widely used signals for imaging are secondary electrons, which are electrons that are excited from the sample molecules by the scanning electron beam. These secondary electrons give information about the texture of the surface and the dark regions in the obtained image mean that secondary electrons are prevented from reaching the detector. There are also other signals obtained when the beam strikes the sample, e.g. backscattered electrons, Auger electrons and X-rays (Zhou et al. 2006). Other methods used to study the morphology of nanocomposites are Atomic Force Microscopy (AFM), Fourier Transform Infrared (FTIR) spectroscopy, etc.

### 27.6.2 Permeability

Permeability is an important parameter for the evaluation of nanocellulose-reinforced composite films for packaging applications. The experiment with diffusion cells is used to determine the permeability of nanocellulose-reinforced film (van den Mooter et al. 1994). The cellulose nanoparticles confer improved barrier

properties to the membranes used for packaging purposes. Due to the nanosize, gas molecules penetrate with difficulty into the crystalline domains of the cellulose nanoparticles. In addition, the cellulosic nanoparticles form a dense percolating network held together by strong inter-particle bonds, which reinforce their use in films. Meanwhile, CNFs have a strong gas barrier property compared to the CNCs. CNC's can form a dense hydrogen bonding network which can increase the gas barrier property of CNC-reinforced composite films. CNFs have tunable barrier properties which can be used to process high performance barrier films for packaging applications.

The mass transport rate of water/gases is expressed by the diffusion of specimens across a membrane according to Fick's first law, assuming that the linear concentration drop within the film (two-film theory). Using a partition coefficient  $K$ , the bulk concentrations in the diffusion cell chambers can be related to the concentrations on the surface of the film. Another assumption that is made is that the concentration of the diffused specimens present within the film is negligible compared to the total concentrations of the chamber. The complete derivation can be seen in the work of (van den Mooter et al. 1994). It is important to note that the permeability is scaled against the film thickness, resulting in the unit  $\text{m}^2/\text{s}$ .

### 27.6.3 Thermal Analysis

Thermal analysis is defined as the measurement of the physical and chemical properties of materials as a function of temperature. The two main thermal analysis techniques are thermogravimetric analysis (TGA) and differential thermal analysis (DTA). TGA is a method of thermal analysis in which changes in the physical and chemical properties of polymer materials are measured as a function of the increase in temperature (with constant heating rate) or as a function of time (with constant temperature and/or constant mass loss). The thermal decomposition property of nanocellulose-reinforced composite films is determined by TGA. The DTA measures the temperature difference between a sample and an inert reference material as a function of temperature. This method detects changes in heat content. Differential Scanning Calorimetry (DSC) is another closely related and modified version of DTA. The study of the thermal behavior of nanocomposites helps to evaluate the operational range of work of the nanocellulose-reinforced polymer composites as compared to the traditional composites. The nanocrystalline cellulose (NCC) reinforcement leads to an improvement in glass-rubber transition temperatures, melting point and thermal stability. Kaushik and Grewal (2011) studied the thermal behavior of the TPS/NCC composites for food packaging applications. The thermal degradation of the nanocomposites with 10% CNCs was studied using TGA under nitrogen atmosphere at a heating rate of  $10\text{ }^\circ\text{C}/\text{min}$ . The authors reported that the addition of CNCs had a significant effect on the activation energy for thermal degradation of the composite materials compared to the net matrix alone.

### 27.6.4 *Dynamic Mechanical Analysis (DMA)*

DMA is a viscoelastic technique that monitors changes in properties due to temperature and/or frequency or time change. The technique measures the total energy stored and dissipated in the material due to a dynamic stimuli. The viscoelastic properties are obtained from elastic and viscous responses. The elastic response is a measure of the energy stored in the material and yields the storage modulus ( $E'$ ). On the other hand, the viscous response measures the energy dissipated in the material due to friction and internal movements, and yields the loss modulus ( $E''$ ) (Barari et al. 2016). The effect of CNF reinforcement on the viscoelastic properties of epoxy composites was examined by Barari et al. (2016) using the DMA.

### 27.6.5 *Differential Scanning Calorimetry (DSC)*

DSC determines the energy changes within a material during the constant heating rate. These energy changes correspond to chemical reactions that occur in the sample or physical changes such as glass transition, crystallization, melting of crystals or sample decomposition (Sandler et al. 1998). The DSC can be considered as another thermal test method to measure the  $T_g$  of nanocellulose-based polymer composite samples. The  $T_g$  indicates the temperature at which polymers transform from hard state (glassy state) to soft (rubbery) state. The  $T_g$  increases with increasing chain stiffness and with increasing intermolecular attraction forces. Gray et al. (2018) studied DSC thermograms of low density polyethylene (LDPE)/TPS nanocomposites reinforced with CNCs. These authors reported that CNC increased the  $T_g$  and melting temperature ( $T_m$ ) of LDPE/TPS blends, specially at low TPS content. This can be attributed to the strong interactions between hydroxyl groups of CNC with TPS and CNC increasing the crystallinity of TPS, shifting its melting point to higher temperatures. The authors also concluded that these LDPE/TPS/CNC composites can be a sustainable alternative to replace LDPE in food packaging application.

### 27.6.6 *Thermal Stability*

TGA is used to determine the thermal decomposition property of nanocellulose-reinforced composite films. The weight loss due to the formation of volatile products after degradation at high temperature is monitored as a function of temperature. When the sample is heated under an inert gas flow, a non-oxidative degradation occurs, while under the flow of air or oxygen an oxidative degradation occurs. In general, the use of clay as filler into the polymer matrix improves thermal stability by acting as a superior insulator and mass transport barrier to the volatile products generated during decomposition (Sinha and Okamoto 2003).

## 27.7 Properties of Polymer Nanocomposites

The composite materials exhibit outstanding mechanical properties with the mixture of nanocellulose and polymer matrix at low filler loading (Dufresne 2012). Mechanical solicitation is achieved within the polymer matrix under suitable condition by mechanically percolating a stiff network of nanoparticles. With the increase in the aspect ratio of CNCs, the stiffness of the percolating CNC increases (Bras et al. 2011), higher aspect ratio of the CNC is more important from the mechanical point of view, since it induces a decrease in the critical percolation threshold and stiffens the continuous network. In case of inhibition of formation of percolating nanoparticle network, only the high stiffness of crystalline cellulose in the nanoscale dimensions, the high aspect ratio and filler-matrix interactions are involved in the reinforcement phenomenon.

### 27.7.1 Optical Properties

The ultraviolet-visible (UV-Vis) spectrometer is used to calculate optical properties such as regular light transmittance (Tr) of nanocellulose-polymer composite films (Fig. 27.11). Fujisawa et al. (2012) reported that TEMPO oxidized cellulose nanofibrils (TOCNs) when reinforced in polystyrene (PS) provides superior reinforcement at low concentrations and improves optical transparency of PS composite film. Savadekar and Mhaske (2012) measured the Tr of the TPS/NCF films with a thickness of 0.60 mm using an UV-Vis spectroscope (UV-160A, Shimadzu, Japan) in a wavelength range of 200–800 nm. Soeta et al. (2015) prepared low-birefringent and highly tough poly(ethylene glycol) (PEG)-grafted CNF/cellulose triacetate (CTA) nanocomposite films. Due to the nanometric size effect of the TOCNs with a uniform width of ~3 nm, the PEG-TOCN/CTA nanocomposite films had a high transparency and a low birefringence with potential use in transparent optical films.

### 27.7.2 Barrier Properties of Nanocomposite Films

The food packaging materials used today are made from non-biodegradable polymeric materials which lead to serious environmental problems such as disposal, recycling etc. The preference for the use of these materials for food packaging polymers is their low cost, ease of processing and excellent barrier properties Lavoine et al. (2012). Biobased nanocellulose-based composites with improved barrier properties are desirable in our society to develop efficient, biodegradable and environmentally friendly packaging materials in the future. However, the low permeability of the polymers can be improved by reinforcing highly crystalline nature nanocellulose particles, since they can form a dense percolating network. In addition, there are strong particle-polymer-polymer molecular interactions, since these nano-sized particles have a greater ability to bond to the surrounding polymer

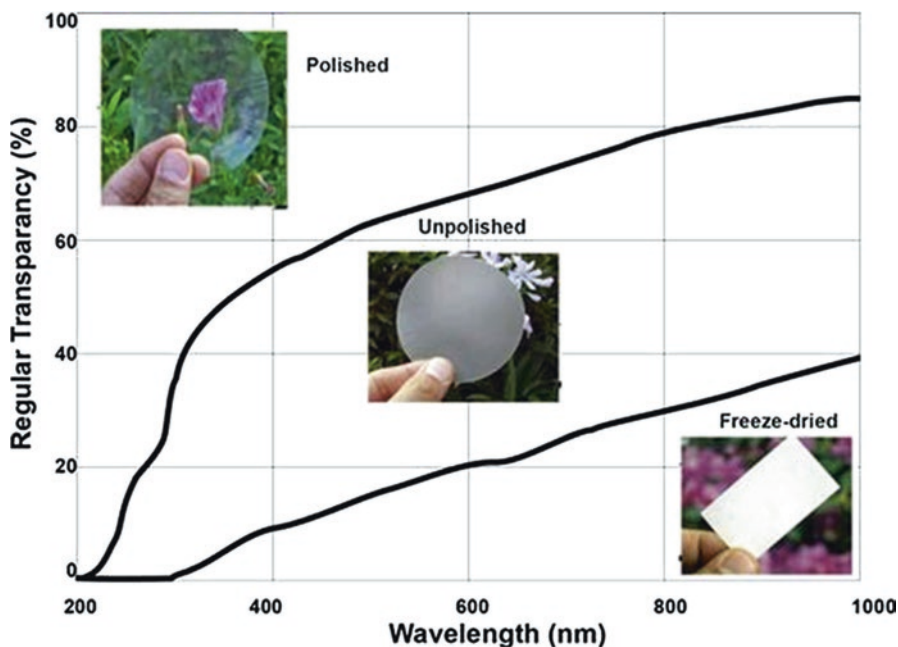


Fig. 27.11 Light transmittance of microfibrillated cellulose films (Lee et al. 2009a)

material, thereby reducing the chain segmental mobility and penetrant diffusivity (Dufresne 2013). Nanocomposite films prepared using nanocellulose as reinforcement prolong the shelf life of food and also improve the food quality, as they can serve as carriers of active substances such as antioxidants and antimicrobials (Andresen et al. 2007).

### 27.7.2.1 Oxygen Transmission Rate (OTR)

The nanocellulose can form a dense network with hydrogen bonds which can increase the gas barrier property of nanocellulose-reinforced composite films. Nanocellulose possesses tunable barrier properties which can be used to process high-performance oxygen barrier films for packaging applications (Nair et al. 2014).

Plackett et al. (2010) observed that the addition of 15 wt. % of CNFs substantially increased the oxygen barrier properties of amylopectin films. Saxena et al. (2010) produced a nanocomposite film with low oxygen permeability by casting an aqueous solution containing xylan, sorbitol and NCC. The oxygen permeability of the films prepared from xylan, sorbitol and 50% by weight of sulfonated CNC exhibited a significantly reduced oxygen permeability of  $0.1799 \text{ cm}^3 \cdot \mu\text{m}^2 \cdot \text{d} \cdot \text{kPa}$  compared with films prepared exclusively from xylan and sorbitol with an oxygen permeability of  $189.16 \text{ cm}^3 \cdot \mu\text{m}^2 \cdot \text{d} \cdot \text{kPa}$ . Savadekar and Mhaske (2012) determined

the OTR of the TPS/NCF films using an oxygen transmission rate (OTR) test machine (Labthink BTY-B1). After placing a film in a cell, the oxygen flow was introduced on one side of the films and the OTR was measured. The OTR in ( $\text{cm}^3/\text{m}^2 \text{ d Pa}$ ) was calculated from the mean OTR multiplied by the film thickness ( $\mu\text{m}$ ) and divided by the oxygen gradient in the cell of the testing machine ( $1 \text{ kgf/cm}^2$ ).

Savadekar et al. (2012) determined the OTR of NFC/kappa-carrageenan (KCRG) composite film using an OTR test machine (Labthink BTY-B1). The authors reported that incorporation of 0.4 wt. % of NFC reinforcement led to reduced oxygen permeability of the KCRG/NFC composite compared to KCRG which has a high oxygen permeability. The incorporation of NFC led to the generation of tortuous path for the permeation of oxygen molecules into the KCRG biopolymer matrix, thus leading to a decrease in the OTR.

The PLA bionanocomposites containing 5 wt% of nanocrystals exhibited the highest oxygen barrier. The OTR for PLA nanocomposites with 5% w/w of unmodified CNCs was  $17.4 \pm 1.4 \text{ cm}^3 \text{ mm m}^{-2} \text{ day}^{-1}$ , while that for CNCs modified with an ethoxylated nonylphenol acid phosphate ester in a 1/4 (wt/wt) the ratio was  $15.8 \pm 0.6 \text{ cm}^3 \text{ mm m}^{-2} \text{ day}^{-1}$  (Fortunati et al. 2012b). The addition of 1 wt% of silver nanoparticles to the modified CNC-PLA composites further decreased the OTR to  $12.6 \pm 0.1 \text{ cm}^3 \text{ mm m}^{-2} \text{ day}^{-1}$  (Fortunati et al. 2013). The OTR values of ternary systems consisting of PLA, PHB (poly hydroxybutyrate) and 5 wt% unmodified CNCs was  $15.3 \text{ cm}^3 \text{ mm m}^{-2} \text{ day}^{-1}$ , while for modified CNCs with an ethoxylated nonylphenol acid phosphate ester in a 1/1 (wt/wt) ratio was  $13 \text{ cm}^3 \text{ mm m}^{-2} \text{ day}^{-1}$  (Arrieta et al. 2014).

### 27.7.2.2 Water Vapor Permeability (WVP)/Moisture Vapor Transmission Rate (MVTR)

The water vapor transmission rate (WVTR) is an important property for packaging materials because it is essential to determine the shelf life of the products in the package and the low WVTR of the packaging material allows longer storage time and shelf life.

Svagan et al. (2009) used a dynamic vapor sorption apparatus from surface measurement systems to determine the water sorption kinetics of nanocellulose-reinforced plasticized starch nanocomposite film. The moisture diffusivity of nanocomposite films decreased rapidly with increasing nanofiber content and the diffusivity of the net cellulose network was comparatively very low. The reinforcement of natural biopolymers with CNCs reduced the WVTR of the resulting bionanocomposites. Saxena and Ragauskas (2009) prepared composite films using 10% sulfonated CNCs as reinforcement in xylan polymer. The authors reported a 74% reduction in the specific water transmission properties compared with the film without CNCs. Khan et al. (2014) reported that WVP decreased considerably in CNC-reinforced methyl cellulose composite films with increased in CNC content. The WVP of the control films (without CNCs) was  $6.3 \text{ g.mm/m}^2.\text{day.kPa}$ , while the

nanocomposite films with 1 wt% CNC showed a WVP decreased of 4.7 g.mm/m<sup>2</sup>. day.kPa. Static WVP of cellulose whisker-reinforced rubber nanocomposite films was determined by Bras et al. (2010) according to the standard. They estimated the moisture sorption test of cellulose whisker-rubber nanocomposite films at a relative humidity (RH) of about 75% at 25 °C using a saturated solution of sodium chloride.

Savadekar and Mhaske (2012) determined the WVTR values of TPS/NCF films gravimetrically according to the ASTM E96 method. Each test film was sealed at the top of the permeation cells containing distilled water. The permeation cells were placed in desiccators maintained at 0% RH. RH 0 was maintained using anhydrous calcium chloride (CaCl<sub>2</sub>) in a cell. The composite films were cut into circles and sealed on the cell with melted paraffin. The water transferred through the film and absorbed by the desiccant was determined from the weight of the permeation cell. CaCl<sub>2</sub> was used as a desiccant. Each permeation cell was weighed in an interval of 24h. The WVTR was expressed in gm mm/cm<sup>2</sup> per day. They observed that WVTR decreased significantly with increasing content of NCF in NCF/TPS films. Seydibeyoglu and Oksman (2008) estimated the WVTR of the KCRG/NFC nanocomposite film and found that the high WVTR of KCRG decreases due to the dispersion of NFC in the KCRG matrix.

Song et al. (2014) conducted the WVTR tests on packaging paper coated with PLA/NCF composite material at 23 °C and 50% RH and at 37.8 °C and 90% RH according to the TAPPI standards. They reported that nanocomposite-coated paper samples showed a lower WVTR compared to packaging paper. Pereira et al. (2014) reinforced CNCs as fillers in PVOH matrix. The addition of 5 wt% CNCs decreased the WVP of pure PVOH films from 0.61 ± 0.04 g.mm/kPa.h.m<sup>2</sup> to 0.44 ± 0.01 g. mm/kPa.h.m<sup>2</sup>. Reddy and Rhim (2014) reported a significant improvement in the water vapor barrier properties of agar composite films when reinforced with crystallized nanocellulose following the standard method of ASTM E96-95 with modification. The nanocellulose reinforcement (0.3 wt. %) lead to an improvement in the WVP of the composite film than pure agar film and, therefore, the resulting nanocomposite films could be used in completely biodegradable food packaging. The moisture vapor transmission rate (MVTR) tests were conducted gravimetrically on nanocellulose-reinforced semi-IPN composite films of PVA/PAAm using an ASTM method (ASTM Method 04.06:E96, 1983) by Mandal and Chakraborty (2015). These authors reported the highest barrier property for composite films with nanocellulose reinforcement of 5 wt. %.

Gray et al. (2018) reported that WVP coefficient and WVTR of the LDPE/TPS blends were greatly reduced by 1% CNC reinforcement, leading to better water vapor barrier properties. The results can be attributed to a good interfacial adhesion between the CNC and the starch which restricts the swelling and moisture diffusion of composites. In addition, the CNC loading at a certain level creates a dense and rigid network with hydrogen bonds which acts as a physical barrier for the transport of diffusing molecules.



### 27.7.3 *Stiffness and Strength Properties*

The stiffness and strength properties of nanocellulose polymer composites depend to a large extent on the shape of the nanocellulose. TS is the stress required to break the polymer material while being stretched. TS of the polymer increases with increasing saturation of the molecular weight at a given value. The nanocellulose upon the reinforcement improves the TS of the polymer composites as compared to traditional fillers. The TS of polymer materials was tested by using an universal testing machine by different researchers, namely Ching et al. (2015) reported that the YM of PVA-nanocellulose composite films increased with the reinforcement of small amount of nanocellulose due to the homogenous distribution of the nanofillers and the high interfacial surface area between the nanocellulose and the PVA matrix. YM is the ratio of stress to the strain in the linearly elastic region of polymer composite. It is a measure of the stiffness of the material and is measured using an Instron. Yakkani et al. (2015) characterized the YM of PP/CNF/Fusabond hybrid composite samples by tensile testing according to the ASTM D638 standard. They observed that there was 87% increase in the YM of PP/CNF composite (1.067 GPa) compared to that of pure polypropylene (0.570 GPa) indicating good dispersion of CNFs in the PP matrix with (0.1 wt %) loading of coupling agent Fusabond.

De Menezes et al. (2009) prepared a nanocomposite film by reinforcing cellulose nanowhiskers (CNWs) in highly hydrophobic semicrystalline commodity plastic LDPE. The nanocellulose whiskers were added at 5, 10 and 15 wt. % and the surface of the cellulose whiskers was chemically modified using organic acid chlorides such as stearoyl chloride. The homogeneity of the nanocomposite increased with the length of the grafted chains. The elongation at break increased when sufficiently long chains were grafted onto the surface of the nanoparticles. The TS of the film decreased by 23%, while the YM increased by 24% with respect to the LDPE when it was reinforced with 5 wt. % of nanocellulose whiskers.

Paralika et al. (2008) observed an increase in TS and toughness in the membranes of the PVA matrix with NCC as a filler. The performance of nanocomposite further improved with the additional addition of poly(acrylic acid) (PAA). Ruiz et al. (2000) prepared NFC epoxy nanocomposites. These authors observed that the nanocomposites had the ability to be associated by means of hydrogen bonds and showed great aspect ratios. The benefits were associated with strong interactions between the epoxy network and the CNCs and the creation of a percolating network linked by hydrogen bonds between the NFC-epoxy. The best results were obtained with a 2% NFC reinforcement in epoxy resin with increased mechanical properties. Nakagaito and Yano (2008) also reported the effect of fiber content on the mechanical and thermal expansion properties of biocomposites based on NFC. A linear increase in YM with a fiber content of up to 40% was observed using a PF resin. The results also showed a correlation between the CTE relative to fiber content,

which indicates the effective reinforcement reached by the NFC. The compression molding method was used to prepare composite with NFC sheets impregnated with phenol formaldehyde. The composites mechanical properties were YM (19 GPa) and bending strength up to 370 MPa.

#### **27.7.4 Thermal Degradation**

Very limited studies have been conducted to study the thermal degradation behavior of MFC-reinforced composite films. The thermal properties depend mainly on the MFC preparation process, as well as on the drying process Lavoine et al. (2012). It has been reported with respect to the first point that TEMPO-oxidized cellulose displays multiple degradation peaks Ruiz et al. (2000). Between 225–231 °C, maximum weight loss occurs (60–80%). Quievy et al. (2010) studied influence of the drying process on the thermal stability of MFC films obtained by homogenization. After freeze drying, the MFC gel formed a mat corresponding to the microfibril aggregates, in which some of the microfibrils remained distinct. The freezing step at 20 °C played an important role in the formation of agglomerates. In addition, this step even changes in the rheological properties of the MFC suspension.

#### **27.7.5 Biodegradability**

The degradability of the composites is important when a polymeric system is applied in daily life, since their degree of weight loss has a direct influence on the environment (Gutiérrez 2018c). The effect of different amounts of NCF on the weight loss rate of the film is done by the burial test in the soil. Different researchers have conducted studies of biodegradation of nanocellulose-reinforced polymer composites, since biodegradability is one of the key factors that influence the use of nanocellulose as a filler in the composites.

Bras et al. (2010) studied the effect of cellulose whiskers on the biodegradation of natural rubber (NR)/cellulose whisker nanocomposites. These authors observed that the cellulose whiskers significantly improved the biodegradation of the NR in the soil. Net NR lost about 19% of its weight after being buried in the soil for 4 weeks, while NR containing 7.5 and 12.5 wt% of cellulose whiskers lost about 62 and 71%, respectively, after the same period buried in the soil. Machado et al. (2012) developed biodegradable packaging using cassava starch plasticized with glycerol and nanocellulose extracted from coconut fiber. During storage, the packaging materials were oxidized, thus serving as a sacrificial material to minimize oxidation of the contents of the package. Abraham et al. (2012) studied the biodegradation of NR reinforced with nanocellulose by the vermi composting method with reference to the crosslinking of the NR matrix. They reported that the weight percentage of reinforced nanocellulose greatly influences the rate of biodegradation

of the nanocomposite. The rate of biodegradation by vermi composting was comparatively higher in non-cross-linked composites than its cross-linked counterpart.

Maiti et al. (2013) studied the biodegradation behavior of PMMA/microcellulose crystalline (MCC) nanocomposites prepared by *in situ* polymerization (IPC) and *ex situ* dispersion method (EPC). Biodegradation studies of IPC and EPC films were performed in a simulated aerobic compost environment for 60 days. IPC showed a greater weight loss compared to the EPC and unreinforced PMMA. Arrieta et al. (2014) studied the degradability of PLA-PHB-CNC composites under composting conditions based on ISO 20200 standard. These authors observed that PLA-CNC and PLA-PHB-CNC lost more than 90% of the initial matter in 10 days; PLA and PLA-PHB-CNC in 14 days and PLA-PHB in 21 days. Luzi et al. (2015) studied the biodegradability of surfactant modified CNC (s-CNC)-reinforced PLA composites. They reported that the nanocomposite films disintegrated in less than 14 days and CNC modified with surfactant promoted the disintegration behavior. They summarized that films may have a potential application for short-term food packaging with low environmental impact. Gois et al. (2018) studied the biodegradation behavior of PLA nanocomposites with net (PLA/CNW) and surfactant modified CNWs (PLA/s-CNW) in garden soil. They reported that addition of surfactant accelerated the biodegradation of nanocomposites films and PLA/CNW/PEG1000 showed the fastest biodegradation rate.

## 27.8 Application of Polymer Nanocomposites in Food Packaging

The nanocellulose during the last few years has been used for the preparation of nanocomposites due to its reinforcing property (Table 27.3) Lavoine et al. (2014). The dimension of nanocellulose and its ability to build a strong entangled nanoporous network has fostered the emergence of high-value applications. Various materials have been cross-linked with nanocellulose by various approaches to obtain multifunctional properties such as improved coloration and dyeing, mechanical properties, barrier properties and WVP. Cs (a natural linear polysaccharide) is the second largest natural polysaccharide available which consists of 1,4-linked 2 amino-deoxy  $\beta$ -D glucan. It is a non-toxic, biodegradable, biofunctional and biocompatible material. Many researchers have reported that it has strong antifungal and antibacterial activities Darmadji and Izumimoto (1994). Films made from Cs have been used successfully as packaging material for the preservation of quality foods (Jo et al. 2001). It was found that 3–5% NCC-loaded Cs films gave the best TS values. The improvement in mechanical properties was mainly due to the formation of a percolating network and stronger filler-matrix interactions. NCC also causes improvement in the barrier properties by reducing the WVP and swelling properties. The study of the surface morphology of nanocomposite films indicated the homogeneous structure due to the adequate dispersion of NCC into the Cs matrix. Therefore, nanocomposite films produced by reinforcement of NCC would

**Table 27.3** Application of nanocellulose based composites and their properties

Polymer	Type of NC	Products	Mechanical properties						Applications	Reference
			TS (MPa)	YM (MPa)	GBP (gm m <sup>-2</sup> day kPa)	EB (%)	WVP (gm m <sup>-2</sup> day)			
Chitosan/ Chitosan Sodium caseinate/ Nanocellulose/ Chitosan	NCC	NCC-chitosan nanocomposite films/ biodegradable films	55.3 — 245 (Increased)	4430 (Increased)	Decreased	47	3.31–2.23/0.23 ± 10.11/12.91	Transparent functional packaging/ edible food packaging nanoreinforcer materials in biodegradable packaging/ edible films for improving shelf life	Azeredo et al. (2010); Dehnad et al. (2014)	
Methylcellulose (MC)	NC	Nanocomposite film	12.6 (Increased)	237.6 (Increased)	—	9.2	10.9 × 10 <sup>13</sup> Kg m/Pa s m <sup>2</sup>	Edible food packaging	Pereda et al. (2010)	
		NC-MC nanocomposite biodegradable films	—	—	50 kGy	—	6.34 (Decreased)	Biodegradable cellulosic based packaging	Khan et al. (2010)	
Polycaprolactone MC matrix	NCC	NCC- reinforced nanocomposite antimicrobial diffusion films	18.7 + 3.7	124.1 + 14.6	—	—	—	Vegetable packages	Boumail et al. (2013)	

Kappa-carrageenan	NFC	NFC-kappa-carrageenan based nanocomposite films	26	—	—	—	—	—	—	—	Food packaging	Savadekar et al. (2012)
Alginate biopolymer/Alginate	Cellulose nanoparticles	Nanocomposite films	18.03–9	480	—	—	13	14	—	—	Food packaging	Abdollahi et al. (2013), (2010)
Kenaf/Corn starch	CNC	CNC–Starch nanocomposite films	3.5–8.2	326.1	—	—	38	8.68	—	—	Biobased packing/Edible packaging	Piermaria et al. (2009)
Glucomannan, Pectin, Gelatin	NC	Nanocomposite films	68	14	—	—	7	3.3	—	—	Food packaging	Stuart et al. (2010)
Hydroxypropyl methylcellulose	MFC	MFC based nanocomposite films	—	—	—	—	—	6.39	—	—	Edible films, Transparent films	Zhu et al. (2011)
Caffeine based paper	MFC	MFC-coated papers	47 ± 4	7.3 ± 0.5	—	—	1.2 ± 0.1	—	—	—	Antimicrobial action for food-packing application	Lavoine et al. (2014)
Vermiculite nanoplatelets + Nanocellulose fiber	Nanocellulose fibers	Biohybrid nanocomposite films	257	17.3	—	—	—	—	—	—	Flexible packaging	Aulin et al. (2012)
Thermoplastic starch/ chitosan	BC or NFC	BC/NFC based nanocomposite films	30	15	—	—	—	—	—	—	Functional packaging	Walker (2012)

(continued)

Table 27.3 (continued)

Polymer	Type of NC	Products	Mechanical properties						Applications	Reference
			TS (MPa)	YM (MPa)	GBP (gm m <sup>-2</sup> day kPa)	EB (%)	WVP (gm m <sup>-2</sup> day)			
Sugarcane bagasse nanofibers	Cellulose nanofibres	Cellulose nanocomposite film	140	12.8	—	12.8	5.2	Barrier and protective film in food packaging	Ghaderi et al. (2014)	
Poly(lactic acid) (PLA)	Cellulose nanofiber	Cellulose nanofiber-PLA nanocomposite films	3.6 GPa	71	—	—	—	Food packaging, water and milk bottles, degradable plastic bags/green-based packaging materials/short-term food packaging	Jonoobi et al. (2010)	
LDPE	CNF	Glossy and transparent nanocomposite films	31	500	—	675	0.099	Film applications, like bread and frozen food bags, flexible lids, squeezable food bottles	Chambi and Grosso (2011)	

HDPE	CNF	Transparent nanocomposite films	27.8	—	—	—	—	0.02	Bottles for milk, juice and water; margarine tubs, cereal box liners	Smith (1986)
PP	CNF	Nanocomposite film	43	1550	—	600	0.06	Thin film coatings/ Yogurt containers and margarine tubs, microwavable packaging	Azeredo et al. (2010)	
PVC	CNF	Nanocomposite film	55	21	—	450	0.2	Films for improving shelf life	Azeredo et al. (2010)	
Pea starch	CNF	Nanocomposite film	45	1584	—	94	66	Edible films for packaging	Azeredo et al. (2010)	
Poly Propylene Carbocate PPC	CNW	Nanocomposite film	26.9	—	—	2960	—	Food packaging	Sainz et al. (2011)	
Poly Vinyl Alcohol	CNF	Nanocomposite film	3.4	—	108.53	—	—	Packaging film	Tome et al. (2013)	
Alginate	CNW	Nanocomposite film	22.4	290	—	8.25	1.64 g/m <sup>2</sup> s Pa	Food packaging	Abdollahi et al. (2013)	

*TS*, tensile strength; *YM*, young's modulus; *GBP*, gas barrier properties; *EB*, elongation at break; *WVP*, water vapor permeability; *NCC*, nanocrystalline cellulose; *NC*, nanocellulose; *NFC*, nanofibrillated cellulose; *CNC*, cellulose nanocrystal; *CNW*, cellulose nanowhisker; *MFC*, microfibrillated cellulose; *HDPE*, high-density polyethylene; *PVC*, polyvinyl chloride; *CNF*, carbon nanofibre

have a promising impact on food packaging due to their excellent barrier and mechanical properties. A higher concentration of cellulose significantly increased the YM and TS of the composite films, but decreased elongation. Antimicrobial diffusion films (ADFs) for food applications were developed by Boumil et al. (2013) which exhibited the highest TS on storage.

Savadekar et al. (2012) also successfully prepared NFC from short staple cotton fibers using a chemo-mechanical process. It was found in this study that it is important to provide an idea of the use and characteristics of NFC in KCRG for the preparation of biocomposite films.

Abdollahi et al. (2013) developed a biobased nanocomposite by incorporating nanocellulose obtained from acid hydrolysis using sulfuric acid into alginate biopolymer using solution casting method. With the increase in nanocellulose content from 0 to 5%, the TS of composite films increased, but with the further increase in the filler content TS decreased. Nevertheless, with the incorporation of nanocellulose, the transparency of the film decreased essentially at a high level (10%), which suggested the occurrence of a partial agglomeration of the fillers at 10% that coincided with mechanical and micro structural results.

A novel protocol to combine vermiculite nanoplatelets with NCF dispersion into functional biohybrid films was proposed by Aulin et al. (2012). The NCFs of 20 nm diameter and several micrometers long were mixed with high aspect ratio exfoliated vermiculite nanoplatelets through HPH. The resultant hybrid films obtained after solvent evaporation were stiff (YM of 17.3 GPa), strong (strength up to 257 MPa) and transparent.

The study of the oxygen barrier properties revealed an oxygen permeability of  $0.07 \text{ cm}^3 \mu\text{m m}^{-2} \text{ d}^{-1} \text{ kPa}^{-1}$  at 50% RH for the biohybrid films. In addition, the addition of nanoclay also significantly improved the water vapor barrier properties of biohybrid films. The excellent oxygen barrier and optical properties of films could be explored in a flexible packaging for oxygen sensitive devices such as thin film transistors or organic light emitting diode displays. It would also be a suitable packaging material for gas storage applications, barrier coatings/laminations etc. PLA is a versatile biodegradable polymer derived from natural sources. It is produced by ROP of lactide or by polycondensation of lactic acid. It is a suitable polymer matrix for the preparation of high-quality composite films by the incorporation of nanocellulose (Tsuji 2005).

Jonoobi et al. (2010) investigated the effect of CNFs in PLA matrix in terms of mechanical properties and dynamic mechanical properties for food packaging applications. The nanocomposite films with 5% CNF showed an increase in TS, YM and a better viscoelastic behavior.

Ghaderi et al. (2014) used cellulose nanofibers extracted from sugar cane bagasse to be employed in all cellulose nanocomposite (ACNC) films. The study showed that a very low value agricultural waste can be converted into a high-performance nanocomposite (TS 140 MPa). It was also found that the WVP of ACNC increased with more dissolution time. Therefore, due to the promising properties of ACNC, such as toughness, biobase, biodegradability and acceptable levels of WVP, they can be considered a multi-performance material with potential for application in the



barrier and protective food packaging industry. The tensile properties of ACNC films are at least comparable or better than other biodegradable or nonbiodegradable films (Ghaderi et al. 2014).

The highest TS of 8.2 MPa was shown by a biocomposite having 6% CNC in thermoplastic cassava starch (TPCS). This suggests the stress transfer and interfacial interactions between the matrix phase and the filler, which is related to the high L/D and efficiency of the fiber treatment. The Kenaf fibers are also found to be compatible with agar and starch made from potato, and films was tested for their potential use in food packaging (Piermaria et al. 2009; Smith 1986).

PLA is used as a food packaging polymer for short-term products such as drinking cups, salad cups, containers, wrappers and laminated films (Sedlarik et al. 2012). But the gas and water vapor properties of the PLA make it insufficient for its use in some cases (Ljungberg and Wesslén 2002). Therefore, the preparation of PLA nanocomposites is a method to improve their properties. Song et al. (2014) prepared novel biodegradable composites by incorporating hydrophobic-modified NCF into a biodegradable PLA matrix. These authors suggested that addition of NCF improves the water vapor barrier property of the PLA matrix. The modified NCF/PLA composite at a concentration of 1 wt. % of NCF when coated on paper by a coating process reduced the WVTR of paper to 34 g/m<sup>2</sup>/d wrt from 1315g/m<sup>2</sup>/d of pure PLA. Paper is a promising green based packaging material. Similar effects on barrier of PLA/cellulose nanowhisiker composites were also reported by Paralikar et al. (2008). Fortunati et al. (2013) prepared nanocomposite films by reinforcing NCC and silver nanoparticles in PLA matrix, which were found to be effective against Gram-positive (*S. aureus*) and Gram-negative (*E. coli*) bacteria and yeast.

Similarly, Reddy and Rhim (2014) prepared completely biodegradable food packaging material using CNCs and agar. The addition of CNCs lead to the improvement of the mechanical properties of the CNC/agar composite as it resulted in increased TS and YM and decreased in WVP.

The edible food packaging films were prepared by Pereda et al. (2010) by reinforcing the nanocellulose in the Cs matrix. Nanocellulose-based Cs composite films for antimicrobial meat packaging applications were prepared in a similar manner by Dehnad et al. (2014) and Khan et al. (2014). Ciesla et al. (2014) prepared PVA-starch-nanocellulose films for food packaging intended for radiation decontamination. They observed that the reinforcement of nanocellulose in starch-PVA films lead to an essential improvement in the mechanical properties of the films and a reduction in irradiation sensitivity as compared to starch-PVA films. They also noted that hydrophilicity of composite films can be reduced by irradiation.

## 27.9 Future Prospects and Challenges

Today, the current trend of packaging materials is that they are ecofriendly, biodegradable and not plastic. But, on the contrary, it is not so easy to avoid plastic due to its inherent quality, particularly in high population countries. However, the western

world countries have initiated commercial production and the use of biobased material for packaging. But the problems are many and the availability of the raw material and supply chain management for a sustainable production is a great obstacle to satisfy the enormous needs of the market. The future packaging can be addressed through the proper management of materials that address environmental concerns, e.g. nano based composite materials, 100% biodegradable packaging, IP, innovative packaging using unutilized agromass residues to reduce carbon footprint etc. The petroleum-based packaging material can also be reinforced with biobased filler to facilitate biodegradation. Appropriate chemistry must be developed to be able to easily resolve the compatibility of the hydrophilic and hydrophobic properties of materials.

Another nearby field in food packaging is the use of stimuli-responsive polymer materials. These novel type of packaging materials can adapt to the surrounding environment and regulate the transport of molecules according to the reaction in response to external stimuli (Buonocore and Iannace 2013). To sustain life and maintain biological function, nature requires selectively tailored molecular assemblies and interfaces that provide a specific chemical function and structure, as well as a change in their environment (Stuart et al. 2010). Nanocellulose reinforcements offer potential advantages in specific properties related to their lower density and other advantages such as low CTE, transparency and barrier properties. NFCs have begun to receive additional attention as reinforcement material due to reductions in energy requirements to break down cellulose fibers to NFC.

Despite the continuous research in nanotechnology, there are many challenges that must be addressed in the future for economic production and, thus, ensure its efficient utilization as a filler/ reinforcement agent in different polymer composites. It is necessary to develop economical and profitable processes for the production of nanocellulose with optimal characteristics. The widespread application of these materials will require additional research to address problems related to their hydrophilic nature in many applications. Appropriate chemistry and techniques are needed to adequately disperse nanocellulose reinforcers or convert them into a useful form for incorporation into a variety of matrices and strongly bond them to it. A more efficient control of the structure at multiple scales is needed to tailor performance. The development of new analytical methods is necessary for the simulation of processing and also for the prediction of the mechanical properties of nanocellulose-based structures. The process modelling effort will be required to link the modelling of NFC distributions to optimize the properties. The appropriate applications requirement must be identified, investigated and demonstrated.

To date, the availability of significant amounts of nanocellulose has impeded faster research and development efforts on a large scale related to its use. If high-value applications can be found, it may be possible to integrate nanocellulose production into the materials flow of these bio-refineries, where it could potentially help improve economic announcements (Zhu et al. 2011) of commercial and government pilot plant-scale production facilities, which will probably improve the situation (Walker 2012).

## 27.10 Conclusion

Today's society is an expert in technology, but at the same time it is also concerned about its surrounding and its future impact. The modernization and sustainable growth that refer to any sector are complimentary to each other and every individual of mother earth wants to make this complimentary bonding stronger.

Due to more advances in composite materials, there is paradigm shift in material science. The packaging material is preferred more natural and composite based instead of monolithic material. Nanomaterials are also equally important to improve the functional character of the packaging material and reduce the material requirement in terms of material savings for sustainable use.

Nanocellulose has an enormous application in food packaging due to its competition with synthetic materials and infinite availability. Functional properties such as physical, chemical and barrier properties in case of nanocellulose composites strongly recommend that it would be the most suitable material for ecofriendly food packaging material in future. The problem remains with the hydrophilic nature of the nanocellulose material and the dispensing capacity with the matrix, which can be addressed by chemical and mechanical action or a combination of both. It is also understood that the nanocellulose composite often outweighs the cost, concern and with time the problems will be further refined and also solve the various problems facing the food packaging industry in present era.

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