Chapter 2
Non–Wooden Cellulose Materials Sourced From Plant Wastes

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ABSTRACT

Cellulose is the most abundant plant waste material, making it a strong candidate to replace petroleum products as a future polymer material. Since cellulose is also abundant in food wastes, upcycling technology to obtain functional materials from these wastes is reviewed from the perspective of resource recycling. Cellulose, which is particularly difficult to handle as a material, has the property of being insoluble in solvents due to its strong cohesive nature. For this reason, recycled cellulose also is discussed as regenerative celluloses. However, compared to these chemically modified celluloses, the utilization of biomass cellulose fibers obtained from plant waste, which are a less scientific process and inexhaustible, enable to contribute to a sustainable society. The current status and technology of unmodified cellulose fibers is presented.Especially, the properties of cellulose hydrogels, which are agglomerated cellulose, and films are introduced in this chapter.

INTRODUCTION

Humanity's social life and profit-seeking economic society have prioritized affluence and we now face the collapse of the global environment and are being forced to take countermeasures. Under these circumstances, we are now opening the door to economic vitality rooted in a low-carbon and recycling-oriented society. But contrary to this trend, plenty of wastes is produced due to the increase activity of agricultural, food and paper industry representing a tremendous challenge for the environment. Mean-

while, industrial waste products are considered as the promising and the suitable material to meet the growing demand for feedstock in terms of resource depletion and conservation of environmental resources (Jawaid & Khalil, 2011). Following this social trend, the conversion of currently underutilized wastes into resources is necessary for the economy to coexist in harmony with nature (Kobayashi, et. al 2022). Reflecting the seriousness of the huge annual disposal of petroleum-based plastics, sustainable green materials are becoming an interesting alternative. Therefore, it is essential to reduce the plastic availability of non-biodegradable petrochemical-based materials, and alternatives such as biomass-based materials are attracting attention. So, it is inevitable that biomass increasingly used in the development of biodegradable films would become value-added bioproducts, and the following approaches can be considered. Natural and non-synthetic polymers are attracting attention to overcome environmental problems, and biomass-derived materials will contribute significantly to a low-carbon society. As a material that can replace synthetic plastics derived from petroleum resources, biomass-derived materials are attracting attention now. Among these sources, as a usual biomass material, polysaccharide is well-known and representative natural polymers like cellulose, which is the most abundant source in the biosphere. In particular, cellulose waste, which has high content of raw materials, is a promising biomass alternative source like food waste and agro-waste. For example, several plants waste of bagasse and agro-waste of bamboo, banana, corn stalks, coir, rice and husk and also oil palm waste are mostly popular sources and the cellulose contents in them are abundant in natural cellulose fiber (Joshi et al., 2004, Jawaid & Abdul Khalil, 2011; Kalia et al., 2009). However, cellulose fiber is found with other materials such as lignin, hemicelluloses and pectin and the percentage in each fiber depend on fiber source, variety, harvest conditions and more (Doree, 1947). For the regenerated cellulose processes from such sources, cellulose has stiffness structure with microfibriles having hydrophilic and hydrophobic parts with complexity structures of the partial crystalline (Swatloski et al., 2002). Therefore, the isolation of cellulose fiber from waste products needs intensive treatment. There are several methods for cellulose obtaining from fibers such as chemical treatments, mechanical treatment, and chemo-mechanical treatment. However, the regenerated cellulose process is limited in dissolving cellulose due to the rigid structure of microfibrils, which have hydrophobic portions and a complex structure of partial crystals (Swatlowski et al., 2002). To date, several studies have been conducted on solvents that dissolve cellulose, especially for solvent systems providing an unstable structure of ether, ester, and acetal derivatives like cellulose acetate, cellulose nitrate, and cellulose xanthate (Heinze & Liebert, 2012). Such regenerated celluloses are fibers that are created by chemically dissolving cellulose and then regenerating it into fibers. These fibers are produced by dissolving cellulose in specific solvents and regenerating by precipitating in an aqueous medium. But there are not many reports on unmodified cellulose (Figure 1). In recent years, there has been a trend to reconsider the use of conventional plastics, which cause oceanic problems such as microplastics (Li, 2007) and require huge amounts of energy to produce, however, this is in contrast to the issue of a low-carbon, recycling-based society. In addition, the recent trend toward energy reduction through lighter weight materials is also driving progress in the study of cellulose-added reinforcing materials. For these reasons, Japan's Ministry of the Environment has been focusing on cellulose nanofibers (hereafter referred to as CNF), a plant-derived material (Japan ministry environment, 2021).

Figure 1. Chemical structure of cellulose
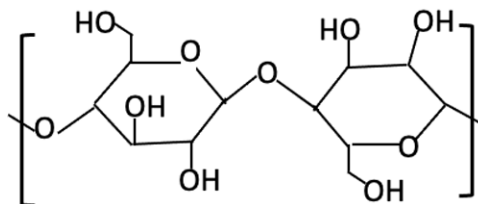
The Ministry of the Environment of Japan is promoting the CNF Performance Evaluation Model Project with the aim of effectively reducing CO_2 emissions through the use of materials that utilize CNF reinforced resin materials (composite resins, etc.) as the foundation of various products, etc., by focusing on CNF (Cai et al. 2014) and micro-crystalline cellulose (MCC) (Majeed et al. 2013). Thus, CNFs and their composites are one of such examples, finding exponentially increasing popularity from a research and development (Kalia, et al. 2011; Trache et al., 2020), but they seem to be still far from being industrially practical at this time.

It has been known that cellulose molecules are natural polymers consisting of linear chains of glucose. Cellulose fiber is a linear, high-molecular-weight homopolysaccharide made up of β-1,4-D-glucose units (Figure 1). Bundles of plant-derived cellulose molecular chains are called cellulose microfibrils (single nanofibers), which have a width of 3-4 nm and are composed of crystalline, associate crystalline, and amorphous portions. In plant tissues, cellulose microfibrils are further bundled into bundles and exist as cellulose fibers of 20 to 40 μm. When cellulose fibers are unraveled to a width of 100 nm or less, which is at the nanoscale, they are called nanocellulose. The technique of defibrillating microfibrils of cellulose fibers at the molecular level is a physical means for nano-cellulose to be insoluble in ordinary solvents. But, in the later year, the direct dissolving process of the regenerated cellulose by using non-derivativing solvents, the cooper salts and concentrated ammonia were effectively cotton dissoluble processes in terms of cuprammonium (Launer & Wilson, 1950) and also alternative ammonia compound as LiCl/DMAc (Striegel, 1997) and metal complexing solvents were used transition metals such as ionic liquid solvents (Heinze & Liebertm, 2012).

On the other hand, sources of cellulose fiber include woody component like pulp and non-woody sources such as agro-products like natural cellulose fibers including cotton and lignin (Batra, S. K, 1985), but the key applications of cellulose fibers include spun yarn, clothing, and fabrics. Among cellulose sources, food waste is currently mostly incinerated or left outdoors after disposal and these food wastes contain less impurities like sugar, lignin, and proteins, and are known to have high cellulose content. Since several approaches to regenerate such wastes to cellulose fibers were reported for scaffolds elaboration from their source, establishment of technology for wastes upcycling toward value-added materials and products becomes important. In these trends using cellulose materials, there is very little movement to convert food waste into ADVANCED MATERIALS and If unused waste and food waste can be utilized as a resource, it is a material that is in line with a sustainable society. For example, the extracted cellulose was used for producing sustainable regenerated textile fibers (Costa et al., 2013). In films, regenerated cellulose fibers attracted more attention to produce the forms used for food and medicine packaging (Klemm et al., 2005) due to their properties of biodegradable, highly transparent, flexible, and low-cost

thin gas barrier (Yang et al., 2013). Also, refining and using food-based waste like agave and sugarcane bagasse (Tovar-Carrillo et al., 2014; Nakasone & Kobayashi, 2016) and thinning bamboo agent (Tovar-Carrillo et al., 2013) were reported as cellulose sources since 2013 (Kobayashi, 2015). Therefore, as considered as renewable organic polymeric source was produced in the biosphere (Wooding, 2001), cellulose is a material with great potential for raw material used in the industry widely. Unfortunately, less than 2% was recovered industrially, representing an important waste against disposal problem. So, the re-use of agro-industrial waste is needed, because of the serious economic and environmental problems caused by disposal of these resources.

General Properties of Various Plant Wastes

In order to evaluate several waste fiber properties many studies have been carried out reporting isolation and characterization (Abdul Khalil et al., 2010; Abba et al., 2013; Namvar et al., 2014). Table 1 shows properties of several waste fibers and Figure 2 shows pictures of plant waste and their SEM images for sugarcane, palm leaf, oil palm empty fruit bunch (EFB), coconut and agave.

Table 1. Properties of some waste fibers

Fiber	Fiber length (mm)	Width fiber (μm)	Diameter (μm)	Density g/cm³	Moisture content (%)	UTS (MPa)	Modulus (GPa)
Cotton	3.00-3.50	60.00-110		1.5		500-880	0.05
Jute	3.00-3.60	70.00-120	200	1.45	12	460-533	2.5-13
Coir	2.00-3.00	14.0-17.65	100-450	1.15	10~12	131-175	4~6
Banana	0.90-4.00	80.00-250.00	80-250	1.35	1-~12	529-754	7.7-20.8
Sisal	0.85-1.00	100-300	50-200	1.45	11	568-640	9.4-15.8
Conifer	3.10-3.60	65.00-120		1.5		1100	100
Kraft fiber	1.25-1.60	19.56-20.10		1.54		1000	40
Sunhemp	0.85-1.00	100-250	48	0.673		200-300	2.68
Pineapple	3.00-9.00	20.00-80.00	20-80	1.44		413-1627	34.5
Palmleaf	1.25-1.74	19.58-23.10	240			98.14	2.22
Kenaf	0.66-0.82	17.70-26.70	200	1.47		157.38	12.62
Kusha grass	0.70-0.80	16.03-21.58	390			150.59	5.69

Figure 2. Pictures of plant waste and their SEM images for sugarcane, palm leaf, EFB, coconut and agave
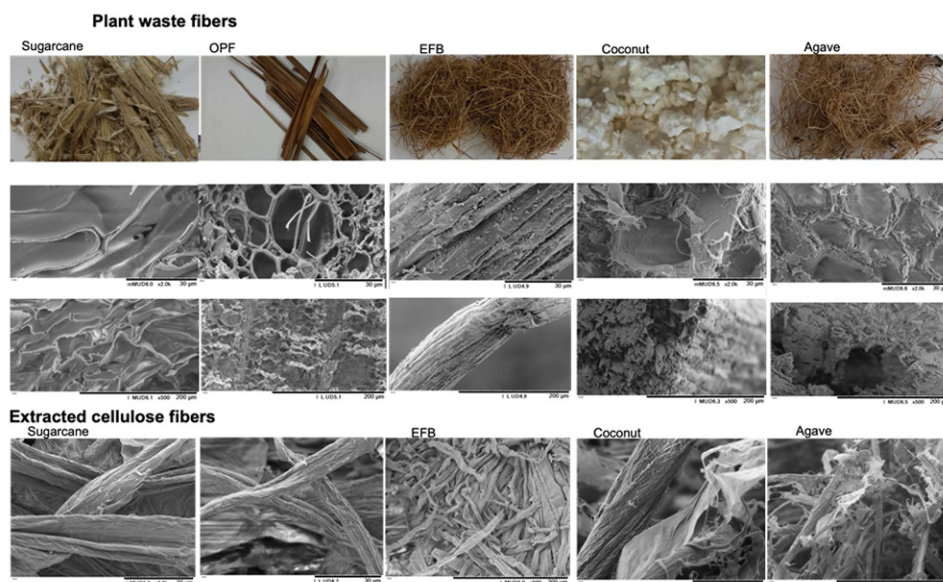
Such waste fibers of plant origin clearly show cellulose microfibrils of about several tens of micrometers, which are aggregates of cellulose molecular chains. The structure differs depending on the waste type. In sugarcane, the cellulose is linearly aggregated, while in agave and EFB, the agglomerated fibrils are already broken up into thick fibers of several hundred μm. This is due to the fact that the fibrillation occurs in high-pressure presses during the squeezing of their extracts. In the fibrous residues remaining after the mechanical chipping in this state, fibrous fibrils are still mixed with impurities such as sugars, lignin, and hemicellulose, and coconut also contains a large amount of oil and fat, which can be observed as slightly flat areas in the SEM photograph.

In contrast, the chemical treatment appears to be enabled to remove such impurities from the cellulose fibers. As seen in SEM pictures, the fiber structure was defibrillated in a twisted form, and eventually, bundles of fine fibers of less than 1 μm were observed to form cellulose fibers of several tens of μm in diameter, consisting of several fibrils that run along the length of the fiber (Kalia et al., 2011; Kalia, et al., 2009; Wooding, 2001).

As summarized in Table I for properties of some waste fibers harvested from different plant source, the structure of cell walls of cellulose, which consist of highly crystalline and regular regions of pure cellulose interspace with small regions of amorphous cellulose. The amorphous cellulose regions usually contain impurities (Kalia et al, 2011; Alberts et al., 2002; Thomas et al, 2011). In addition, the bundles of these cellulose ribbons are called microfibrils. The microfibrils generally vary in width from 0.008 to 0.03 μm and the microfibril bundles are held together by hydrogen bonds formed between the hydroxyl groups of the glucose molecules (Thomas et al., 2011). In the SEM images, extracted cellulose fibers are visible as microfibrils. In the plant wall, the main structural unit of cellulose consists of cellulose microfibrils bonded together by polymer resins like other segments of lignin and hemicellulose. The microfibrils are covalently bonded together by various polymeric sugars and proteins forming fibrils with a diameter around 0.05-0.3 μm and length of 0.15 μm to 20 cm approximately (Wooding, 2001; Nair,

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2007; Siro, et. al., 2010). Fibril length and composition is determined by plant species and function as well as waste source for a specific renewable product (Rowell et al., 2000; Satyanarayana, et. al., 2009). The plant cells grow first by forming primary or outer wall after the nucleus has divided during cell division (Osorio et al., 2010). This primary cell wall consists of layered cellulose fibrils. In wood cells, the primary cell wall is formed of matted fibrils resembling matted felt and is usually coated with an outer protective or bonding layer (Batra, 1985; Bledzki & Gassan, 1999). In cotton linters, in contrast to this, the primary wall consists of dense spiral wrapped layers of cellulose fibrils (Azwa et al., 2013) having a waxy cuticle to protect the exposed cell wall. The cotton fibers consist of single individual row of plant cells which are coated by intercellular materials that connect adjoining cell walls together (Cristaldi et al, 2010; Huang et al, 2012). Thus, cotton linters possess a highly defined spiral wrapped pattern of fibrils while the fibrils in wood pulp fibers, which consist mainly of mixture of mature xylem cell, appear almost parallel.

Non-wooden cellulose fibers in their applications.

Since the early 1900s, the need for environment friendly products has been increasing. This has led to a strong trend toward replacing petroleum products with bio-based materials to which the carbon cycle can be adapted. Thus, the use of bio-based polymers, composites, and CNFs, for example, has been gaining momentum. Traditionally, wood pulp has supported the paper industry, which has grown into a large industry (Bajpai, 2014; Reddy, et. al., 2006). However, the supply of wood pulp raw materials is limited, and there is a shift toward the use of recycled pulp. In addition, attempts have been made to convert non-wood pulp into paper (Pardo et al., 2014; Nguong, 2013; Monteiro, et. al., 2011). This is because of the need to protect the global environment from the perspective of resource conservation, non-wood paper is being reevaluated and utilized for various applications. Therefore, utilization of non-wooded materials in the production of paper is one of the most cost-effective and environmentally friendly technologies. In Japan, for example, non-wood paper has been produced and used in the form of Japanese paper since the introduction of paper manufacturing in the United States. Currently, non-wood fibers such as Manila hemp, sisal, flax, and cotton are used for special printing paper such as banknotes, certificates, and certificates, as well as for cigarette paper, teabag paper, electrolytic condenser paper, and various types of filter paper. Non-wood fibers are generally longer and have a greater cell wall thickness than hardwood fibers, making them low-density and bulky, and when made into paper, they have high air permeability (Hara, 1998). In terms of paper strength, such paper has high tearing strength and high strength, and can be used at thinning and low beating. Such non-wooden cellulose fibers offer several properties suitable for diverse industrial applications, for example, in most fibers tensile strengths ranging from 80 MPa for sisal to 938 MPa for ramie. Fiber semi-finished products from non-wood raw materials are widely used in the production of microcrystalline cellulose and cellulose esters in writing, printing, cigarettes, packing, filtering, packaging, sanitary ware, box paper, and more.

Currently, kenaf, ramie, roselle, and flax seed in their polymer matrix composite forms have been studied for their mechanical properties with potential applications in auto parts, mechanical gears, sockets for prostheses, and eco-friendly brakes. The good tensile strength of jute fibers and hemp fibers was utilized in structural applications, construction, sports, domestic products, and auto parts to absorb noises (Abdul Khalil et al., 2010, Abba et al., 2013, Namvar et al., 2014).

Cotton stalks are another important source of raw materials. Cotton is cultivated primarily for textile fibers, and little use is made of the cotton plant stalk. Stalk harvest yield tends to be low and storage can be a problem (Abba et al., 2013; Abdul, el. Al., 2008; Reddy et, al., 2005). Another disadvantage of cotton stalks utilization is uneven distribution of fibers along the stalk induces variable density areas and low resistance to biodegradable agents. The obtained materials exhibited good biocompatibility and bio-integration in the body for cellulose ester (Edgar et al., 2001; Methacanon, et. al., 2010). In the last decade, several studies have reported use of cellulosic natural fibers as a reinforcing agent in biocomposite thermoplastic matrices. Coir, banana, and sisal wastes were used as reinforced polymer composites (Saheb & Jog, 1999, Li et al. 2007). Moreover, the corn husk also was used for biodegradable film, coconut fibers as heat insulator, rice husk in concrete, rice straw and bagasse fiber used as writing and printing papers, and more.

Chemically re-generative treatment of cellulose fibers.

Cellulose is the major component in plants and is an its waste products are excellent for manufacturing many products like regenerate cellulose products of rayon or viscose called commonly, which natural cellulose can be regenerated by xanthogenic acid treatment (Kobayashi et al., 2022; Dungani, et. al., 2014). These are colorless, non-toxic, pure cellulose materials, first trade name cellophane. The raw materials of regenerated cellulose are generally cotton cellulose and wood cellulose, which undergo different manufacturing processes. The process of regeneration is the dissolution treatment of cellulose from solid to solution form, using xanthogenic acid (HS-C(=S) -O-C$_2$H$_5$). Such solvation involves the interaction of the hydroxyl groups of the polysaccharide chains of the cellulose with the solvent, which causes the cellulose fibers dissolve. This allowed cellulose to be converted to the xanthogenated form with carbon disulfide in NaOH. The xanthogenated cellulose solution dissolves in the NaOH solution, resulting in a viscous solution as a cellulose solution (Olsson & Westman, 2013; Liu, et. al., 2011; Lloyd, et. al., 1998).

Cellulose solvents are commonly divided in two main categories of derivatizing and non- derivatizing solvents following the key purpose for regenerated cellulose processes (Kobayashi et al., 2023). For the derivatizing process, cellulose is firstly modified example of cellulose acetate, whose process in acetylation changes the hydroxyl groups to acetic anhydride (Sayyed et al., 2019; Lim, et.al., 2021). Historically, the derivatizing process came first with the covalent modification idea. In 1846, cellulose nitrate was reported to prepare a xyloidine by treating starch, sawdust, and cotton with nitric acid. It becomes the nitrocellulose which can be dissolved in ether and ethyl alcohol (Heinze & Liebert, 2012). Viscous also was prepared via dissolution in caustic soda (Wedin et al., 2018). All of these are made from wood pulp or non-wood pulp but high-purity cotton pulp. On the other hand, in the case of bagasse such as plant waste and food waste sticks shown in Figure 2, chemical treatment methods need to be cellulose fiber first.

In order to obtain cellulose from waste fibers chemical treatments are first step. Chemical treatment has been extensively used to remove non-cellulosic compounds in natural fiber, unfortunately this can affect physical and mechanical of natural fibers, implementing with chemical treatment using chlorite bleaching, alkali treatment and acid hydrolysis (Roberts, 1997).

Moreover, approaches on cellulose fibers from rice husk and from jute cellulose (Jahan et al., 2011), have been reported. Examples for the chemical treatments were reported below. The chemical treatment for preparing cellulosic fibers from cellulose plant fibers used sulphuric acid (H$_2$SO$_4$) and hydrochloric

acid (HCl) were carried out. Also obtaining of cellulose from nanofibers have been reported (Qua et al. 2011; Brinchi et al, 2013). They observed influence on the surface charge and dimensions of the cellulose nanofibers due to chemical treatment with acid hydrolysis. Meanwhile, cellulose fibers were obtained by wet chemical process (Leitner et al. 2007) and, the use of oxygen and peroxide compounds was to remove lignin and other non-cellulosic compounds without degradation of cellulose fibers (Kopania et al, 2012).

Figure 3. Agave fibers treated with different concentrations of NaOCl (Tovar-Carrillo, et al., 2014) and their DMAc/LiCL solution
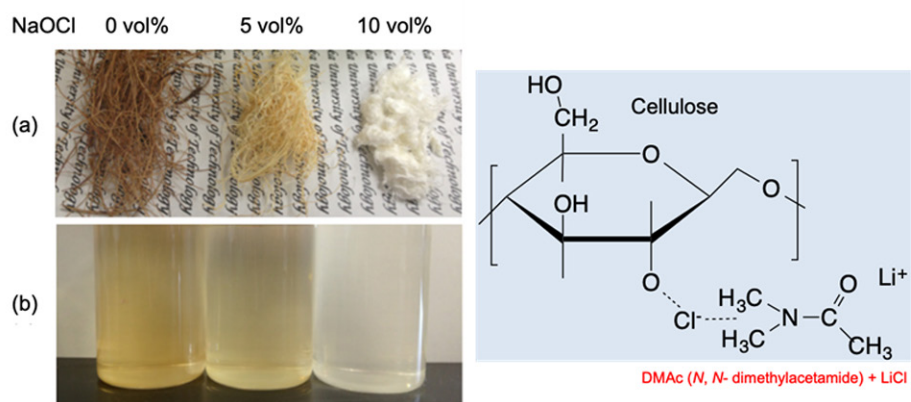
In the cases of food bagasse, several approaches were reported for chemical treatment of agave waste fibers (Tovar-Carrilloet al.,2014), in which lignin was dissolved under alkaline, neutral, or acidic conditions (Nakasone & Kobayashi, 2016b). In the case of acidic method acid sulfite was used with conditions of sulfur dioxide and water at high temperature (Roberts, 1997). The delignification process produced depolymerized cellulose fibers due to acid hydrolysis of glycosidic linkages. Another method consists of alkaline delignification by mixture of sodium sulfide and sodium hydroxide, diminishing crystalline structure of the waste fiber and improving the efficiency of the de-lignification process. During this alkaline process black liquor rich in lignin was obtained and light brown color fibers, bleaching process caused residual lignin from fibers (Figure 3), when used with sodium hypochlorite (NaOCl) (Tovar-Carrillo et al., 2014). In the case of agave bagasse, as shown in Figure 3, the bleaching process with NaOCl changed the color from brown and ochre to white as the amount of bleaching agent increased. After acid and alkali treatment, the non-bleached fiber samples remained slightly brown. The infrared spectra showed that the lignin peaks in the sugarcane sample were almost completely eliminated by this chemical treatment. After the bleaching reaction with chlorine bleach, the sample became white in $NaClO_2$ and gave almost the same spectra as cotton cellulose.

Figure 4 Chemical treatment flow from sugar cane bagasse to cellulose fibers using several chlorine bleaches of NaOCl, NaClO$_2$ and NaClO$_3$ and FT-IR spectra. Sulfuric acid hydrolysis was carried out at 60°C for 1.5 hours, followed by alkali treatment at 10 Wt/V% for 12 hours at 80°C. The resulting non-bleached fibers were then treated with chlorine bleach for 50 hours at 50°C for 3 hours
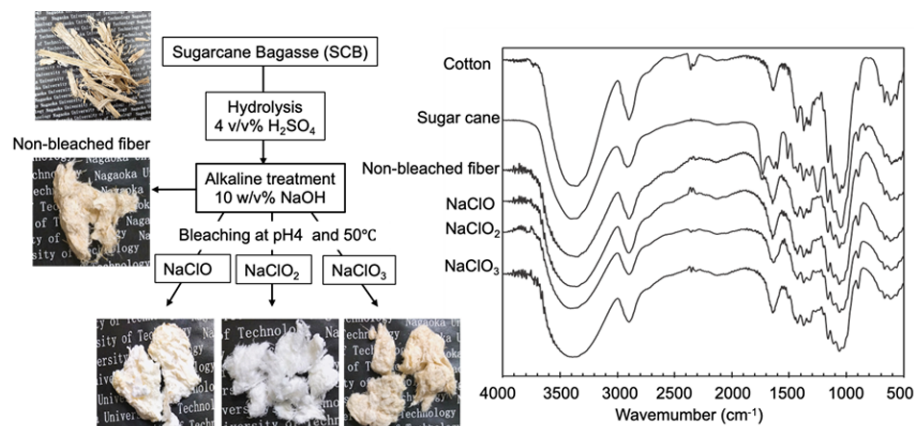
Industrial applications of waste fibers.

Recently, due to necessity of environmentally friendly materials, natural fibers plant waste products offers suitable properties for several industries. The increment of industrial waste products several alternative uses were reported (Izani et al, 2012 & Thiruchitrambalam et al, 2009). Furniture industry increased use of natural fibers as an alternative manufacturing and waste fibers were used for the manufacture of biocomposite advanced (Abdul-Khalil et al, 2012), for example, flax fibers can be used for furniture applications. In other reports, plant fibers such as kenaf, banana, jute and flax has been used as reinforcement. In the last decade, several studies reported use of cellulosic natural fibers as a reinforcing agent in biocomposite thermoplastic matrices; Coir, banana and sisal wastes used as reinforced polymer composites (Saheb & Jog, 1999, Li et al. 2007). Moreover, the corn husk was used for biodegradable film, coconut fibers as heat insulator, rice husk in concrete, rice straw and bagasse fiber used as writing and printing papers, and more.

In the case of automobile industry, waste fibers are the prime candidate for automotive industries. In has been found that by using waste fibers the car weight decreases around 30% and that positive impact on fuel consumption. Recently, biocomposite of polymers reinforced with fibers for manufacturing of seat back, side and door panel, hat rack, boot lining, spare tire lining, business table, dashboard, piller cover panel, instrumental panel and headliner panel have been used (Suddell & Evans, 2005; Bledzki et al, 2006; Holbery & Houston, 2006) among the companies that use those fibers are, Audi, BMW, Fiat, Ford, Renault, Saab, Volvo, Mitsubishi and Peugeot. In addition, Mitsubishi was developed door trim from bamboo composite (Suddell and Evans, 2005; Bledzki et al, 2006; Abdul Khalil, 2012).

Moreover, use of waste fibers as reinforcement of composite materials for packaging increased in the last years. The obtained materials cannot replace completely the properties of synthetic polymers, due to their characteristic hydrophilic behave including permeability to gases and vapor and poor mechani-

cal properties (Johansson et al, 2012; Hirvikorpi et al, 2011 & Siro & Plackett, 2010). However, waste fibers-based materials offers environmental friendly alternative due to their recyclability (Johanson et al, 2012; Majeed et al, 2013). In the last years, several countries have been used waste fibers as composite materials for building applications (Kamble & Behera, 2021; Shakir et al., 2023). Materials such as, doors, panels, roofing sheets and door frames have been elaborated with jute, sisal, and coir. These composite materials produced good insolation and alternative option in low cost. In addition, it has been possible to obtain high transparency films using natural fibers.

Cellulose fibers re-generated from plant wastes

Cellulose solubility. Cellulose solubility is crucial for the obtaining of cellulose materials. Several approaches reported solvent systems for cellulose. Cellulose is hard to dissolve due its stiffness with strong hydrogen bonds linked network. The solvent must be capable to interact with the hydroxyl groups in cellulose fibers, eliminated partially the strong inter-molecular hydrogen-bonding between the polymer chains (Wooding, 2001). However, appropriate solvents systems for cellulose solution obtaining are a key point in order to preserve cellulose fibers properties (Striegel, 1997; Wooding). Table 2 shows several conventional solvent systems for cellulose fibers. Cellulose solubility was reported in acidic condition using sulfuric acid hydrolysis, sulfuric acid destroys the amorphous regions of cellulose fibers (Lindma, et. al, 2010).

Table 2. Conventional and new cellulose solvents

Solvents	Methods and condition	Cellulose	References
NaOH (8-10%)	Direct dissolution 4°C	Treated cellulose, DP 330	Kamide et al. (1984)
NaOH (7-9%)	Direct dissolution	Treated cellulose, DP 330	Yamane et al. (1996)
NaOH (8-9%)	Freeze-thaw	MCC, DP 200	Isogai & Atalla (1998)
NaOH 6%/Urea12%	Freeze-thaw	Cotton linter DP=690	Zhou & Zhang (2000)
NaOH 7%/Urea 12%	Direct dissolution -10°C	Cotton linter DP=700	Cai & Zhang (2005)
NaOH9.5%/ Thiourea 4.5%	Direct dissolution at -4 °C	Cotton linter DP=620	Ruan et al (2004)
NaOH 9%/PEG 1%	Freeze-thaw	Cellulose powder DP=810	Yan & Gao (2008)
NaOH 12-18% Thiourea 4-6%	Two-step, -2 to 5°C	Cotton linter DP=570 Avicel DP 570	Qi et al (2011)
NMMO	Two-step, 90°C	Cotton linter DP=690	Hon (1997)
DMAc/LiCl	Two-step, room temp	Cotton linter DP=700	Striegel (1997)
DMAc/LiCl	Solvent exchange with water, ethanol, DMAc and DMAc/LiCl	Agave bagasse cellulose Sugar cane bagasse cellulose Commercial cotton products	Tovar-Carrillo et al., 2014 Nakasone et al., 2016 Noguchi and Kobayashi, 2020

DP is degree of polymerization.

On the other hand, alkali systems for cellulose solubility including LiOH and NaOH were reported, although some limitations of NaOH-based aqueous systems were found on dissolving wood pulps fibers (Lindma, et al, 2010; Hon, 1997). In order to obtain cellulose solution, effective destruction of intermolecular hydrogen bonding is essential and critical point for cellulose applications, knowing that intermolecular hydrogen bonding in cellular fiber can be broken by using urea (Tovar-Carrillo, 2014). Unfortunately, urea and NaOH systems could diminish cellulose molecular weight due to the impact on braking intermolecular hydrogen bonding of cellulose structure to allow enhancement water solubility. Alternative method for cellulose solubility was known by using lithium chloride (LiCl)/ *N, N*-dimethylacetamide (DMAc) system (Striegel, 1997). This solvent system has the capability of dissolving cellulose with high polymeric degree like cotton fibers. In LiCl/DMAc system, the activation step is a critical point to allow contact of the solvents with cellulose structure and destroy the hydrogen bonding in the polymer chain by exchange solvent process. In the first step slowed of cellulose fibers in water to lose cellulose structure. The inter-and-intramolecular hydrogen bonds are replaced by hydrogen bonds with water. On the second step, DMAc impedes the re-forming of the inter-and-intramolecular bonds. The solvent system works as solvation of cellulose fibers. Finally, after the activation step, cellulose fibers are ready to dissolve in LiCl/DMAc. It was reported that, concentration of LiCl around 2 and 12% was suitable for cellulose optimal dissolution of cotton and pulp fibers (Tovar-Carrillo et al., 2014). In contrast, LiCl concentrations higher than 12% (12%-15%) results in supersaturated solution and cellulose tended to precipitate. It was found that LiCl concentration in LiCl/DMAc solvent system promote different cellulose fiber arrangements. At lower LiCl concentration the fiber arrangement is mainly lineal, and with the increment of LiCl concentration aggregates shapes were reported (Striegel, 1997; Tovar Carrillo, et al, 2013). In this case, various dissolution mechanisms for cellulose in DMAc/LiCl were proposed as following: 1) $[DMAc_n + Li]^+$ macrocation must exist. 2) In the ion cluster with cellulose, the Cl^- anion is dissociated from the Li^+ cation by intercalating with one or more DMAc molecules. 3) The Li^+ cation interacts with the carbonyl group oxygen of the DMAc molecules. 4) The Cl^- anion disrupting the hydrogen bonds of cellulose can create hydrogen-bond-type interactions with the hydroxyl group hydrogens of cellulose. 5) The macrocation must have weak interactions with the cellulose oxygen. But these processes should be mentioned to be no conclusive evidence. Until today, the interactions between Li^+ cation and the glycosidic oxygen were indicated in the solution system described though semi-empirical MNDO computer models. This type of interaction between cation and various disaccharides in the gas phase (Striegel, 1997; Tovar-Carrillo, 2014).

Film fabrication. Films made from waste-derived cellulose fibers are a much-needed alternative to petroleum-derived plastic films. Regenerated cellulose films prepared from several solutions for example, aqueous alkali/urea solutions exhibited good optical transparency and oxygen barrier properties under dry conditions (Yang et al., 2013). Moreover, amorphous cellulose film was dissolved in regenerated from LiCl (8wt%)/DMAc solution when acetone was coagulated. Cellophane is the mostly produced to cellulose-based film, applying for food packaging due to its transparent, strength and flexibility, making it an attractive natural-based material for food wrapping and adhesive tapping and the derivatives made from wood, cotton, or other sources. This film was obtained by chemically treated processes in alkali solution as called mercerization process (Jewkes et al., 1969) and then, the alkali pulp was continually treated with carbon disulfide to convert to an orange solution known as viscous or cellulose xanthate. To reconvert the viscous solution into cellulose, dilute sulfuric acid and sodium sulfate were prepared in a bath for passing the solution (Venkateshwaran, et. al., 2012; Ververis,et. al., 2004). Various applications

were typically coated as nitrocellulose-based waterproof coatings and polyvinylidene chloride-based heat resistance (Wahlang, 2012).

Gelation for cellulose hydrogel films. It was known that cellulose was soluble in DMAc/LiCl, but subsequent material adjustments had to wait until the advent of hydrogel films in 2013 (Tovar-Carrillo et al., 2013). Since cellulose in cellulose-DMAc/LiCl solution (Figure 3) is insoluble in alcohol and water, the presence of these vapors increases the concentration of alcohol and water in the DMAc, causing cellulose to agglomerate and precipitate. A result of such phase inversion from liquid to solid swelling product, hydrogel could be obtained by washing the precipitated transparent gelatinous films having DMAc/LiCl and water (Nakasone and Kobayashi, 2016; Kobayashi, 2018).

Figure 5. Cellulose hydrogel formation via phase inversion process from cellulose liquid solution to gelatinous films. Pictures were hydrogel film of cellulose fibers treated without and with NaOCl bleached agent
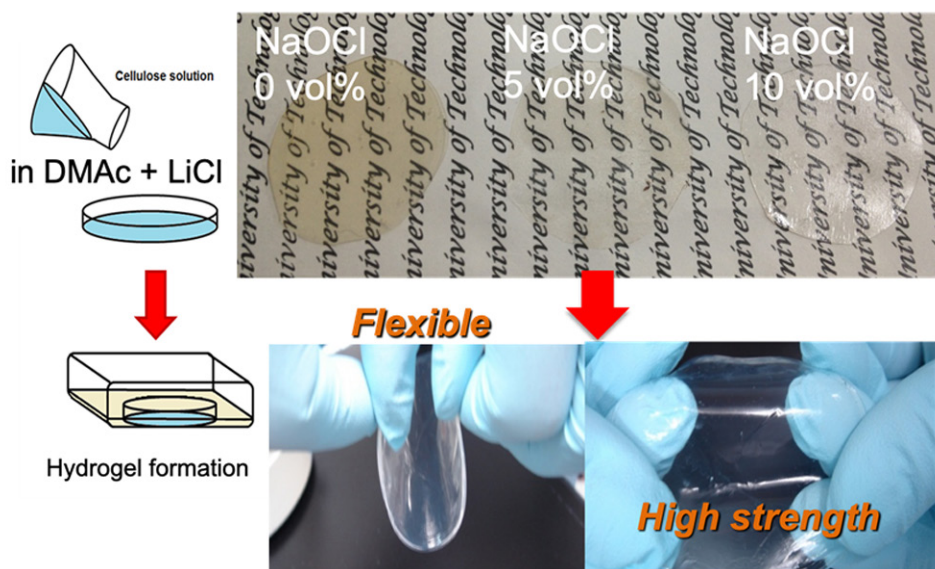

Figure 6 UV-visible absorption spectrum of a film made by heating hydrogel films obtained from NaOCl-treated fibers (as seen in Figure 5) at 150°C-200°C and a photographs of the cellulose films. The absorption spectrum showed that UV light at wavelengths below 350 nm was absorbed by about 60-80%, preventing transmission, while visible light had good transmission performance. The transparency of the cellulose films was ensured, although the film's appearance tended to become slightly cloudy as the heating temperature increased.

44

Figure 6. Ultraviolet-visible region transmittance spectra of cellulosic films heated at 150°C, 180°C and 200°C applied for hydrogel films and their appearance pictures
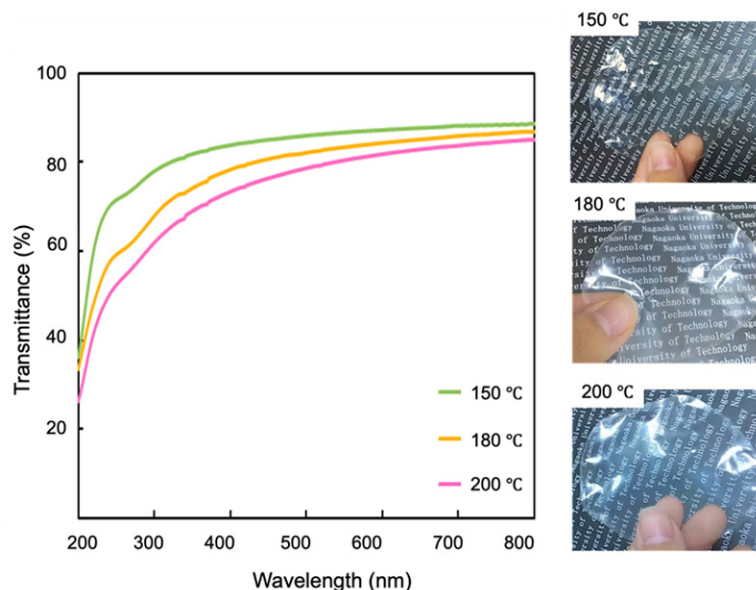
Applications of medical materials elaborated with cellulose obtained from waste fibers.

The main requirement for any material with potential metical application is exhibit long term usage within the body with any adverse or toxic reaction. The material has to be biocompatible for this purpose. Fortunately, the obtained cellulose materials exhibited good biocompatibility and bio-integration in the body for cellulose ester (Edgar et al., 2000). Therefore, cellulose is a suitable candidate to elaborate materials for medical applications due to its nontoxic properties and stable chemically and mechanical as well as completely insoluble under physiological conditions (Bouchard et al. 2006; Dourado et al., 1999) and cellulose polymers are known to have good biocompatibility and wound healing properties like other natural polymers such as chitin, chitosan and alginates (Kimura & Kondo, 2002).

On the other hand, the application of natural polymers for medical applications is limited due to their poor mechanical properties and in some cases, since it is necessary elaborate composite materials combining biopolymers and synthetic polymers (Jiang et al., 2006 ; Henniges et al., 2012). In addition, several materials including cellulose membranes were known to have anti-inflammatory and anti-cancer effects, as reported that hyaluronic acid-carboxymethylcellulose membranes were applied topically for reducing scar formation and surgery (Kim et al., 2013; Kim et al., 2005). Moreover, cellulose membrane has been recognized as a permeation enhancer for drug delivery systems offering a suitable patch material for topical formulations.

Due to cellulose membrane chemical characteristics, it is considered for capsule based controlled drug delivery. Cellulose membranes offered several advantages over conventional capsule materials for uses of controlled release of bulk drug in short time from the membrane (Park et al., 2011; Thombe et al., 1999; Frisbee et al., 2002). Moreover, since the capsule properties were independently modulated without interacting with the core formulation, the obtained capsule was suitable for drug molecules that were difficult and expensive to elaborate, especially sensitive to aqueous environments and elevated temperatures (Kim et al., 2013 ; Kim et al., 2015). But, this provided a smooth, slippery, easily swallowable, and tasteless shell for drug delivery, and these capsules made from regenerated cellulose are commonly used in the commercial fabrication market. Furthermore, cellulosic scaffolds were used for tissue engineering and other medical applications (Digenis et al., 1994; Dahl et al., 1991) and an ideal matrix for large-scale affinity purification procedures (Bussemer & Bodmeier, 2003; Pina & Sousa, 2002), showing chemically inert matrix with excellent physical properties as well as low affinity for nonspecific protein binding suitable for cell adhesion and tissue regeneration.

Cellulose hydrogels for tissue regeneration.

In the last years, several approaches have been conducted to the development of novel hydrogels for tissue regeneration (Langer and Vacanti, 1993) by using natural polymers as elaborated scaffolds for tissue engineering (Sionkowske, 2011; Chen, et al., 2008; Lu et al., 2017). Another reason is that hydrogels are materials that retain large amounts of water, just like living organisms.

Table 3 compares common natural polymers used as scaffolds materials.

Table 3. Summary of commonly employed natural polymers used as scaffolds materials

Main polymer	Properties	Function in wounds	References
Alginate	Wound dressing, Ease application, biodegradability	Wound promotion, Infection control, Haemostatic effect	(Mobed-Miremadi et. Al 2016) Raguvaran et al. 2017)
Chitosan	Wound dressing, In situ gelation, Biodegradability	Healing promotion, Infection control, Anti-inflammatory effect, Haemostatic effect, Delivery of active molecules, Monitoring healing progress	(Balakrishnan et al. 2005, Rocasalbas et. Al. 2013)
Gelatin	In situ gelation, Biodegradability	Delivery of active molecules, Tissue reinforcement, Cell recruitment	(Lee et al. 2014, Jrudi et al. 2015)
Collagen	Wound dressing, In situ gelation, Biodegradability	Delivery of active molecules, Tissue reinforcement, Cell support	(Ribairo et al. 2013, Basu et al. 2017)
Dextran	Skin substitute	Healing promotion, Delivery of active molecules	(Goh et al 2016)
Cellulose	Moisture control	Healing promotion, Cell support	(Wang et al. 2016)
Heparin	Hydrogel sheet, Moisture control	Delivery of active molecules	(Wang et al. 2016)
Glycosaminoglycans	Film dressing, moisture control	Infection control, Tissue reinforcement, Guided tissue regeneration, Promotion of angiogenesis	(Wang et al. 2016, Lu et al. 2017)
Mixed extracellular matrix biopolymers	Sprayable elastic adhesive	Wound closure, Delivery of antimicrobial peptides	(Catanzo et al. 2015, Annabi et al. 2017)

Among them, one strategy has been working with cellulosic polymers, since cellulose natural polymer has easily assimilated in the body due to biocompatibility, hydrophilicity, and biodegradability (Kobayashi & Tovar-Carrillo, 2015). The ultimate purpose of tissue engineering is to replace, repair or enhance the biological function or damage of an organ or tissue. Engineered tissues are produced by using cells that are manipulated through their extracellular environment to allow new tissue formation (Tamada & Ikada, 1993). There are three ways in tissue engineering (Svensson et al. 2005) as followed;

(1) inducing migration of tissue regeneration,
(2) using to encapsulate cells and acting as immuno-isolation barrier,
(3) using as a matrix to support cell growth and cell organization.

Ideals materials of scaffolds should have complex requirements such as no-toxic, biodegradability, appropriate porous structure and mechanical properties. The main characteristic of scaffolds materials is to mimic the extracellular matrix of tissues in the body, both providing support to the cells and having mechanical properties to the tissue. The complex interaction of cells with the extracellular matrix and with neighboring cells is regulated by several reactions. Moreover, extracellular matrix is crucial for cell proliferation, differentiation and death and mediate regeneration and healing process. Base on this, scaffolds materials should provide a stable suitable material for long- term tissue formation without any adverse reaction into the body. Cells do not interact with materials surface directly (Langer, et al. 2006). Cells attach to the absorbed proteins in the scaffold and are important for tissue regeneration.

The implanted material should mimic the environment of the implanted site. After implantation, the first key point is the attachment of water molecules, enhancing protein binding and adsorption (Sionkowske, 2011) and becoming key point of cell adhesion and proliferation, due to cells interact with the adsorbed proteins promoting tissue regeneration. So, hydrogels are suitable scaffolds materials for tissue regeneration for their three dimensional structure. Several approaches have been reported results on tissue regeneration using cellulose hydrogels. Cellulose hydrogels obtained from agave bagasse fibers showed good cyto and biocompatible properties (Tovar-Carrillo, 2013). In addition, cellulose hydrogels exhibited higher cell adhesion number comparing to commercial cell culture assays (Nakasone et al., 2016). Cells adhered to cellulose hydrogels, showing boundaries tightly long shape on the material surface on the first 4 h of cell culture assay (Kobayashi et al., 2015). In the case of skin injuries, severe loss of tissues occurs and scaffolds materials are suitable to intended regenerated tissue due to their three-dimensional structures (Kirker et al., 2002; Raguvaran et al., 2017). Since scaffolds mimic extracellular matrix environment with adequate mechanical and chemical conditions to cell adhesion maintain tissue regeneration for long-termthose, the contribution to mechanical and structural integrity of the surrounded tissue allows nutrients and water transport and facilitated vascularization providing guidance for cell proliferation. In fact, mechanical, strength and stiffness were the most important properties, as well as surface and microstructure (Song et al., 2012; Mishra et al., 2017).

Moreover, scaffolds porosity requirements are dictated by the final application. Pores shape, size and arrangement are important factors to consider. Furthermore, scaffold architecture has an influence on water molecules contact, protein adsorption, cell-cell contact, cell adhesion necessary for cell proliferation and extracellular matrix transport (Gupta, et al., 2008). One important point is cell seeding viability on the scaffold base and source of nutrients and its vascularization ability after implantation to allow cell migration. Scaffolds can be implanted without cells when they are intended to be colonized by cells in short time. Scaffold implantation is determined by surrounded tissue reaction to it, this is mainly a

surface manner (Langer et al., 2006; Brumkar et al., 2006). Moreover, protein adsorption is sensitive to mechanical stiffness. Stiffness allows diffusion and permeability of water molecules important for protein adsorption (Amin, et al. 2014). Surface topography also plays an important role on protein adsorption. Nano and micro roughness increase protein adsorption promoting cell adhesion (Tovar-Carrillo, et al., 2013). During the conditioning of the cellulose hydrogel, varying the LiCl in the DMAc changed the cohesiveness of the cellulose segments in the hydrogel, with cellulose oriented side by side at 6 wt% LiCl and a randomly entangled gel at 12 wt% (Figure 7). The growth of fibroblast cells was examined, and it was found that at 6 wt%, the cells grew in the direction of cellulose fiber cohesion, as if they were oriented, and the number of fibroblast cells grew in such a way that they were oriented along the scaffold fibers.

Figure 7. AFM Phase-shift images (upper) of the agave hydrogel films prepared with (a) 6wt% and (b) 12 wt% of LiCl contained DMAc solution and phase-contrast light images (bottom) of their films of (c) 6wt% and (d) 12 wt%, respectively, for 48h in fibroblast cell culture times (Tovar-Carrillo et al., 2013)
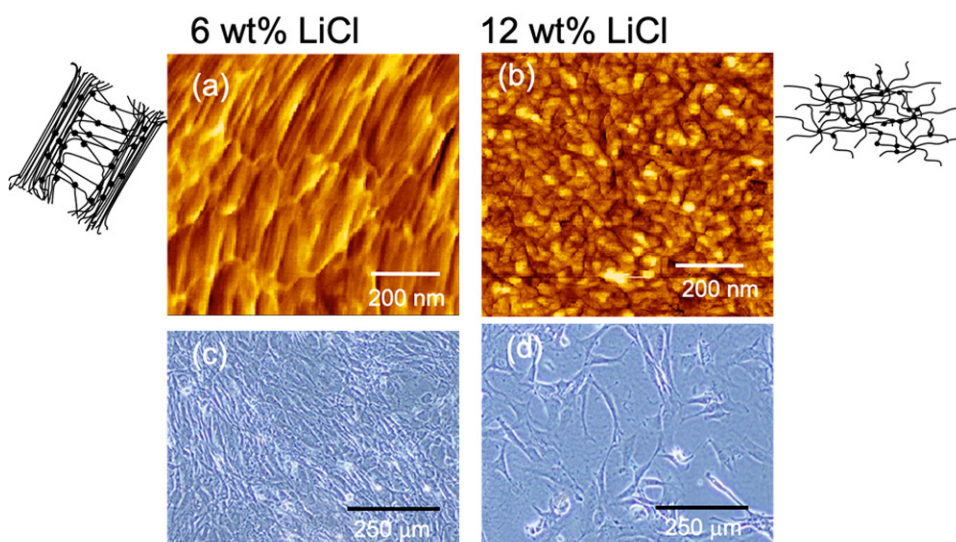

For medical applications, hydrogel and films have been widely used in several fields, such as bio-materials, agriculture, food, and water purification, drug delivery, sensors and smart materials (Gibson, 2005). In pharmaceutical, cellulose has several properties to offer, such as excellent compaction when its blended with other pharmaceutical excipients suitable for oral administration. Cellulose films offered several potentials advantages as a drug delivery excipient (Chen, et al. 2008; Jiang et al., 2016; Iresha & Kobayashi, 2021) and since cellulose films is a low cost, its use provides a substantial environmental advantage compared with other films. Biocompatibility of cellulose hydrogel films regenerated from sugar cane bagasse waste was evaluated by investigating it's *in vivo* behavior in mice (Nakasone at al., 2016). The cellulose hydrogel films were implanted in the intraperitoneal of mice for 4 weeks, showing small influence of the implanted hydrogel films on the growth of mice. It was seen that no inflamma-

tion reaction in the intraperitoneal was observed by post-mortem examination, indicating that cellulosic hydrogel films had excellent in biocompatibility.

In drug delivery field, the incorporation of a second component into the film will change the structure and morphology of the network controlling their diffusion properties. It has been reported that cellulose hydrogels provide space and support for cell adhesion and growing, and potential control and function of the engineered tissue in situ, such as; cartilage, bone, muscle, skin, adipose, artery, ligament, tendon, liver, and bladder (Langer & Vacanti, 1993). In addition, cellulose hydrogels can be used in several applications, including drug delivery systems (Sionkowske, 2011), wound healing (Lloyd, et al. 1998). Cellulosic medicines had excellent response to ultrasound trigger to releasing the embedded drug, when ultrasound was exposed to the hydrogel medicines (Figure 8) (Kobayashi, 2023). The enhanced releasing drug by ultrasound was due to soften effect of cellulose hydrogels by breakage hydrogen bonds in the hydrogel under the exposure, but when the ultrasound stopped, the gel fomes self-healing to be original form. Such hydrogel softening and reforming were observed with sono-deviced rheometer (Noguchi & Kobayashi, 2020), causing by hydrogen bond destruction in gels and reforming in the presence and absence of ultrasound.

Figure 8. Illustration of ultrasound response effect of cellulosic hydrogel. Under ultrasound exposure gelation network destroys by hydrogen bond breakage, but, when the exposure stops, the bonds form again
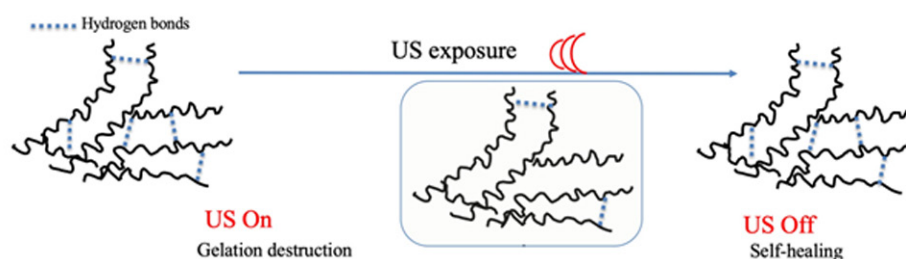

Applications development in current and future.

Materials elaborated from waste fibers can be applied in several fields and industry. In the last years cellulosic materials became a suitable eco-friendly alternative. Regarding to medical field and tissue engineering, cellulose hydrogels are common in non-toxic, water absorbable and reproducibly obtaining flexible. In tissue regeneration, the material interacting with skin cells and enhancing tissue growing at the surface with immunologically inert. It has been found that when the obtaining of the scaffold involves several solvents and compounds traces could remain in the material affecting the biocompatibility of the scaffold. Moreover, cellulosic materials are suitable to implanting in the body (Chandy & Sharma, 1998). In addition, biodegradable nature of cellulose materials became popular, and this property contributes to the construction of a sustainable society as a recyclable material. For example, cellulosic materials can be converted to fertilizer after use. For example, recently, a water-soluble sponge (Figure 9) made from waste cardboard with a binder of agarose was applied to hydroponics. The lettuce cultivation yielded 53

g weight in 42 days, almost equal to the 54 g yield of the commercial urethane sponge. After harvesting, the waste sponge was aerobically fermented in the presence of microorganisms and could be composted.

Figure 9. Hydroponic application and microbial decomposition of cardboard sponges Pictures of lettuce grown on cardboard sponges for 42 days and composted cardboard sponge in microbial presence after harvest
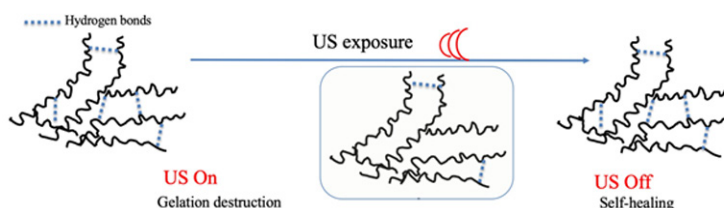
On the other hand, excellent biocompatibility is a practical achievement. In cellulose hydrogel cases, *in vivo* implantation of the hydrogel film in rat model provided excellent results (Nakasone, et al. 2016), showing that non-adverse or inflammation reaction in the peritoneal region after 4 weeks of implantation of the hydrogel film and no cell agglomeration was observed near the hydrogel material. This is because that such cellulose hydrogel is not toxic to the body and showed bio-integration and non-adverse reaction. Several approaches reported results of hydrogels as wound dressings, exudates absorb and moisture retaining for wound healing. In addition, cellulose hydrogels can be applied to solve other needs by regenerative medicines, such as alternative treatments for periodontal and orthodontic, so using for dialyzer membranes for kidney failure (Misha et al. 2017, Meza-valle et al. 2020). Cellulose materials were used as sensing matrix responding to target stimulus, such as pH sensor (Gupta el at. 2008), ammonia sensors (Balakrishnan et al. 2006) and so on. Dialyzer cellulose membranes have the lowest degree of complement with leukocyte activation, inflammatory reactions, and blunting of response of leukocytes (Svensson, et al. 2005), offering an economic alternative for the treatment of this condition. In addition, cellulosic materials had vast applications in medicine detection as sensors (Chen, et al. 2008), pH sensors immobilizing dye on cellulose materials (Svensson, et al. 2005), ammonia sensors (Gibson, 2005), temperature sensors and chemsensors (Sionkowske, 2011). Cellulose materials obtained from waste fibers have a wide number of possibilities on several industrial and medical applications.

CONCLUSION

Due to the growing importance of building a society of sustainability, cellulosic materials made from plant waste have been applied in several industrial sectors in the past few years. In addition, the use of cellulosic fibers, which are abundant as an alternative to petroleum-based plastics, and their industrial diffusion are important factors. In this chapter, cellulose fibers from non-wood plant wastes and un-utilized agro-materials are classified as the main raw materials for obtaining cellulosic materials, and their properties are introduced. The current regenerated cellulose industry was introduced, as well as several papers on the problems of unmodified cellulose fibers and their potential for regenerated cellu-

lose materials. As a distinctive characteristic, the non-toxicity, biocompatibility, and degradability of the obtained cellulosic fibers are described, as well as their economical production, industrialization of recycled cellulose, and environmentally friendly alternative materials. In these cases, cellulosic fibers, have abilities for strong contribution to the future development as advanced materials. We also discussed the economic production and industrialization of recycled cellulose, as well as environmentally friendly alternative materials such as cellulose composites, hydrogels, and films using cellulosic fibers extracted and purified from non-wooden sources.

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