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Cellulose and its derivatives as biodegradable materials: A review

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Abstract

From ancient times to the present day, human beings have carried out many studies to make their lives easier. As a result of these studies, they aimed to discover something new. With the industrial revolution, petrochemical materials began to be produced. The fact that these materials have gone through various chemical stages has become a threat to human health. In addition, the fact that petroleum-derived materials remain in nature for centuries without degrading, and the parallel increase in environmental problems has led humanity to search for different resources. In this work, general information is given about the types of derivatives obtained from cellulose, which is the most common in nature.

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Keywords: Cellulose; Cellulose Derivatives; Cellulose Ether Derivatives; Cellulose Ester Derivatives

1. Cellulose

Plants synthesize cellulose, a polymer that forms in a linear structure [1-4]. Currently, it stands as the most prevalent polysaccharide globally, serving as the primary reservoir of sustainable polymers [2, 5, 6, 7, 8]. Cellulose forms the fundamental structure of both wood and cotton, comprising a substantial 94% of cotton fibers. The wood of trees contains a high proportion of cellulose, exceeding 50%, while its lignin content is comparatively low. Bacteria are capable of producing cellulose, which is referred to as microbial or bacterial cellulose. In contrast to cellulose sourced

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from cotton and wood, which is produced through photosynthesis, bacterial cellulose is not formed through this process and is obtained in a more purified form. [9-10].

Cellulose is comprised of numerous monosaccharide molecules [11]. The arrangements of these monosaccharide units shape the structure of cellulose. Cellulose molecules are created by linking anhydroglucose units, with the chemical formula $(C_6H_{10}O_5)_n$, in a linear chain. The number of anhydroglucose units is referred to as the degree of polymerization (DP, typically averaging between 10,000 and 15,000). These units are linked by 1,4- β -glucosidic bonds, with one of the two anhydroglucose units rotated 180 degrees in relation to the other. The arrangement of cellulose is depicted in Figure 1. As a result, a tension-free linear cellulose structure emerges [1, 3, 6, 7, 12, 13, 14].

Each D-anhydroglucopyranose unit that makes up the cellulose molecule contains hydroxyl groups at the C2, C3, and C6 positions, which can participate in typical reactions of primary and secondary alcohols. Because of its configuration, cellulose 1 can exhibit various characteristics including hydrophobicity, chirality, degradability, and chemical diversity. These collectives establish numerous hydrogen bonds with hydroxyl groups in neighboring chains. The complex network of hydrogen bonds both within and between cellulose chains provides the polymer with stability, increasing the stiffness of cellulose fibrils along their axes. The chains are also stacked regularly in places where the crystal zone will form. In amorphous regions, they form a dispersed stacking. These properties were effective in the formation of the current properties of cellulose [9, 13, 14, 15, 16, 17].

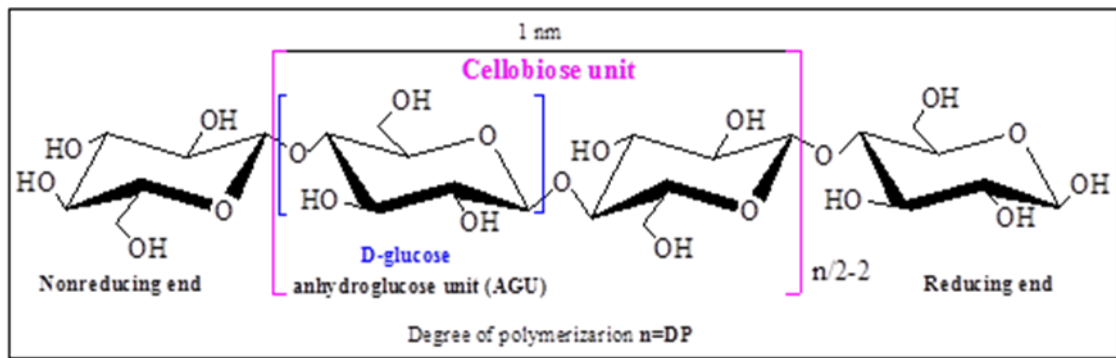


Fig 1. Structure of the cellulose molecule [6].

Cellulose, a polymeric raw material, is primarily utilized in two main fields: the first is the production of materials derived from wood and cotton, and the second is the manufacture of paper and cardboard. Furthermore, cellulose serves as a primary substrate for chemical transformations in the production of cellulose-derived goods, which find utility across diverse fields. The cellulose molecule has been subjected to various reactions to obtain these products, which we call cellulose derivatives. The most important duo of these reactions is etherification and esterification [9, 13, 18].

2. Cellulose ether derivatives

Cellulose ethers are macromolecules derived from cellulose in which the hydroxyl groups of the hydrogen atoms in the anhydroglucose units are replaced by alkyl or substituted alkyl groups. The assessment of key properties of cellulose ethers, such as their molecular weight, chemical composition, distribution of substituent groups, degree of substitution, and molar substitution characteristics, depends on their specific application. These characteristics typically encompass dissolvability, fluid thickness in solutions, surface behavior, qualities of thermoplastic films, and resistance to decomposition by organisms, high temperatures, water breakdown, and oxygen exposure. The viscosity of cellulose ether solutions increases in proportion to their molecular weight. Cellulose ethers, frequently used in

various applications, include methyl cellulose (MC), ethyl cellulose (EC), hydroxyethyl cellulose (HEC), hydroxypropyl cellulose (HPC), hydroxypropylmethyl cellulose (HPMC), carboxymethyl cellulose (CMC), and sodium carboxymethyl cellulose [18-20]. Despite the wide variety of cellulose derivatives produced, The primary factors behind the excessive use of cellulose ethers worldwide are their availability in the market, economic functionality, ease of use, and almost no toxic effects. Therefore, there is a constant expansion in its use. Cellulose ethers offer functionalities such as flow regulation, stabilization, moisture retention, and the formation of protective films. Combining properties such as these cannot generally be achieved when fully synthetic polymers are used [21]. Etherification of cellulose is given in Figure 2

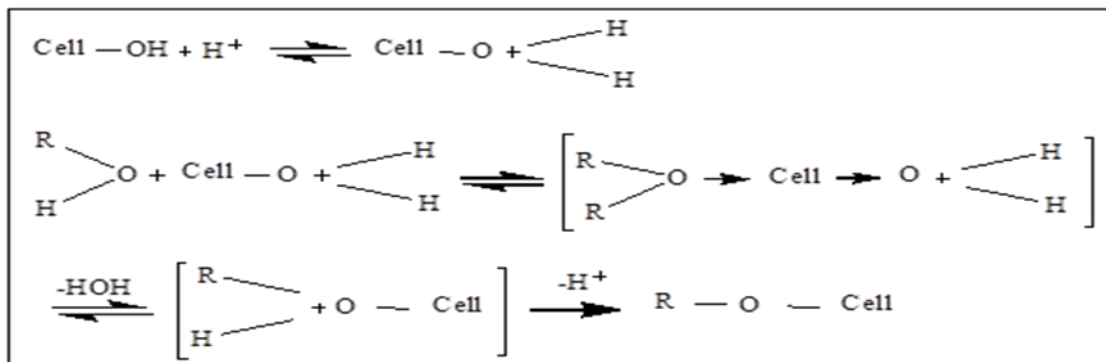


Fig 2. Etherification reaction of cellulose [12].

2.1. Methyl cellulose (MC)

Methyl cellulose (MC) is a key cellulose derivative extensively used in numerous industrial fields due to its diverse applications [22]. MC, was first synthesized as a result of Suida's studies, and it was first named as cellulose ether in 1905 [23]. Methyl cellulose, which is abundant in nature and is expected to stand out as an alternative raw material in the future thanks to its low cost and environmentally friendly properties, is similar to cotton with its neutral, odourless, tasteless and inert structure. MC, is not digested and also has no toxic effects. It swells in water and becomes a light color, making the colloidal solution clear and insoluble in most common organic solvents. It's possible to weaken it by mixing it with ethanol. MC solutions maintain their stability across a broad pH spectrum, from 2 to 12, with negligible alterations in viscosity [11, 24, 25, 26].

MC obtained using iodomethane, chloromethane and dimethyl sulfate is synthesized using the Williamson etherification method. The degree of displacement (DS) of methyl cellulose varies according to its usage areas and purposes. The DS value is determined by the average count of hydroxyl groups (OH) on the anhydroglucose unit that have undergone reaction with the desired substitute group. Since each anhydroglucose unit in the cellulose molecule contains three hydroxyl groups, the highest degree of substitution possible is three, represented as DS = 3 [27].

Methyl cellulose (MC) is extensively utilized as a binder or thickening agent in pharmaceutical chemistry, the food industry, and ceramic processing. It is also employed in electrochemical supercapacitors and electrolyte applications. Methyl cellulose finds extensive application in the food and pharmaceutical sectors owing to its remarkable attributes like superior film-forming capacity, effectiveness as a barrier against lipids, and its ability to minimize permeability to oxygen and moisture vapor [23, 26, 28, 29, 30]. When placed in water, it forms a gel between 60 and 80 degrees Celsius, which dissolves upon cooling. MC is a cellulose derivative with extended chains, encompassing roughly 27 to 32% of hydroxyl groups within its methyl ether framework. The level of polymerization in different classes of

methyl cellulose is between 50-1000 and their molecular weight (average number) is between 10.000 and 220.000 [31-32].

MC undergoes heat-recoverable gelation in aqueous solution by heating in water, and it is also a polymer soluble in water. Cellulose, the base component of methyl cellulose, exhibits an affinity for water. Cellulose fibers resist dissolution in water due to the presence of structured crystalline regions formed through both intra- and intermolecular hydrogen bonding. The formation of crystalline regions in cellulose varies according to its source, yet when a specific quantity of hydroxyl groups undergo substitution with methoxyl groups, it disrupts certain hydrogen bonds, resulting in the solubility of methyl cellulose in water. This change is defined as the number of methoxyl groups separated by the numbers of glucose units and is expressed as the level of substitution (DS). If the degree of displacement (DS) is very low, a sufficient number of hydrogen bonds ensure that methyl cellulose remains undissolved, whereas methyl celluloses with a high degree of displacement (DS) have a hydrophobic and at the same time insoluble structure. Therefore, DS values of products to be used commercially are selected as moderate (1.7-2.0). At colder temperatures, water molecules arrange themselves into "cage-like" formations that encase the hydrophobic methoxyl groups, leading to the solubility of methyl cellulose in water. Above the specified temperature values, these structures deteriorate and break to reveal hydrophobic structures. This phenomenon causes clustered structures to form. Therefore, the gelation phenomenon shows that the hydrophobic effect occurs and also reduces the gel temperature in common solvents that are easily dissolved in water, such as strong electrolytes [25, 30, 33]. The molecular configuration of methyl cellulose is depicted in Figure 3.

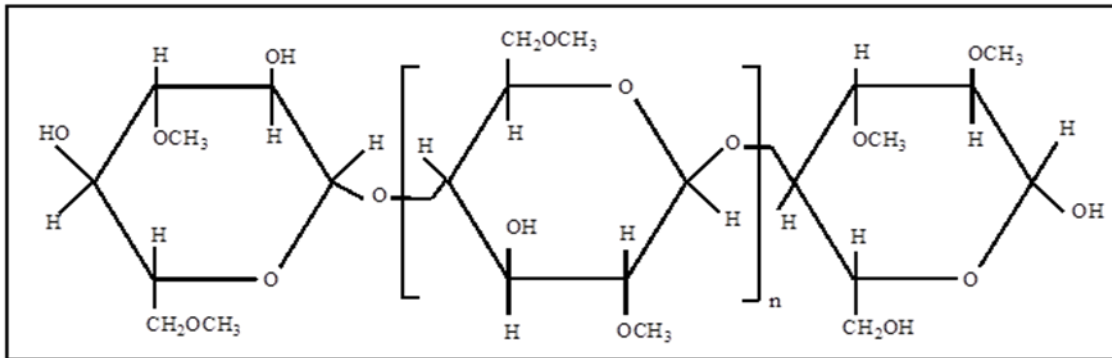


Fig 3. Molecular structure of methyl cellulose [34].

2.2. Ethyl cellulose (EC)

Ethyl cellulose (main source cotton and wood), which is widely used in the pharmaceutical industry [35-36], contains three reactive OH sites in repeating anhydroglyucose units. Some of these hydroxyl groups can react partially or completely with various chemicals, leading to the formation of different cellulose derivatives. As a result of these reactions, ethyl cellulose (one of the most expensive cellulose derivatives), which is obtained by converting cellulose into ethyl ether groups called non-ionic ethyl ether, is partially ethylated and has an ethoxy content ranging from 44% to 51%. It is also a non-ionic and pH insensitive cellulose ether [24, 37, 38, 39]. Thanks to its versatile properties, ethyl cellulose is widely used in optical materials, film forming, packaging, coating material, tablet binder and microencapsulation. We can exemplify these properties as follows [38, 40, 41, 42]: (1) it is white to light skin colored and is in the form of a tasteless, odorless powder or granular material; (2) the melting point is in the range of 240-255°C; (3) its specific density is between 1.07-1.18, its degradation point is between 135 and 155°C, and its combustion temperature is between 330-360°C; (4) insoluble in water but dissolves in various organic solvents, such as ether, ketone, alcohol, and ester; (5) biocompatible, working well with the majority of celluloses, resins, and

plasticizers; (6) is not biodegradable, so it can only be used orally; (7) It is resistant to light, heat, oxygen, moisture and chemicals [40] (8) it is non-toxic [43-44]; (9) has no irritating properties; (10) used as a mold binder to impart plasticity to the flow of particles; (11) possesses the capacity to take pressure, so it protects the coating from breaking during compression [40]; (12) It does not swell or dissolve in water, but it does dissolve in numerous organic solvents [24], so the density and porosity of ethyl cellulose play a key role in drug release from such hydrophobic substances [45]; (13) Despite its lack of solubility in water, it can absorb water. The interaction results from the attraction between water molecules and hydrogen atoms, influenced by the difference in polarity between the oxygen atom and the ethyl group in ethyl cellulose [45-46]; (14) the viscosity of ethyl cellulose varies based on the length of its chains, polymerization degree, or the quantity of anhydroglucose units. At the same time, existing viscosity can also be considered an indirect measure of molecular weight [40]; (15) The glass transition temperature is 120°C; (16) It is physiologically inert, its pH level is between 3 and 11 and does not change. The degree of substitution (DS) for its solubility in water is in the range of 1-1.5, while in organic solvents this value is in the range of 2.4-2.5 [11, 47]. Figure 4 depicts the molecular arrangement of ethyl cellulose.

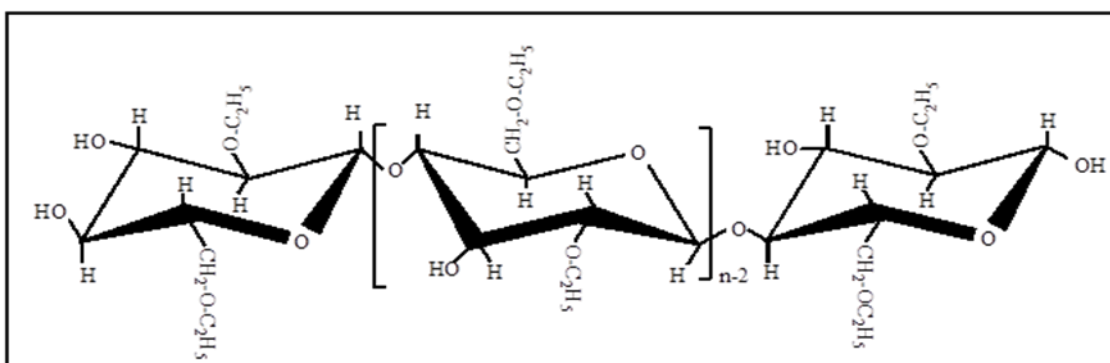


Fig 4. Molecular structure of ethyl cellulose [40].

2.3. Hydroxyethyl cellulose (HEC)

Hydroxyethyl cellulose (HEC) is among the most commonly utilized cellulose derivatives and is a water-soluble and low-density hydrophilic polymer obtained by etherification of alkaline cellulose with ethylene oxide or ethylene chlorohydrin, replacing the H atoms in cellulose with hydroxyethyl groups. Cationic polymers, especially biopolymers, are used as stabilisers and thickeners in paint mixtures and hair and eye care products. With its greatest hydrophilic property, hydroxyethyl cellulose is the most often utilized cellulose ether. Together with other non-ionic, protective colloidal qualities, this soluble cellulose ether has the ability to thicken, suspend, disperse, emulsify, create films, and protect against water. HEC is widely used in many industrial fields such as polymer chemistry, cosmetics, agriculture, building materials, textiles and paper, paints, aerogel production, synthetic resins, ceramic industry, etc [11, 48, 49, 50, 51, 52, 53, 54, 55]. HEC powder can have a high viscosity in aqueous media, which varies depending on temperature and shear rate, and shows pseudoplastic behaviour. Aqueous solutions of hydroxyethyl cellulose are highly viscous and their viscosity varies depending on DS and molecular weight. It is also insoluble in most organic solvents [31, 56]. Figure 5 illustrates the molecular configuration of hydroxyethyl cellulose.

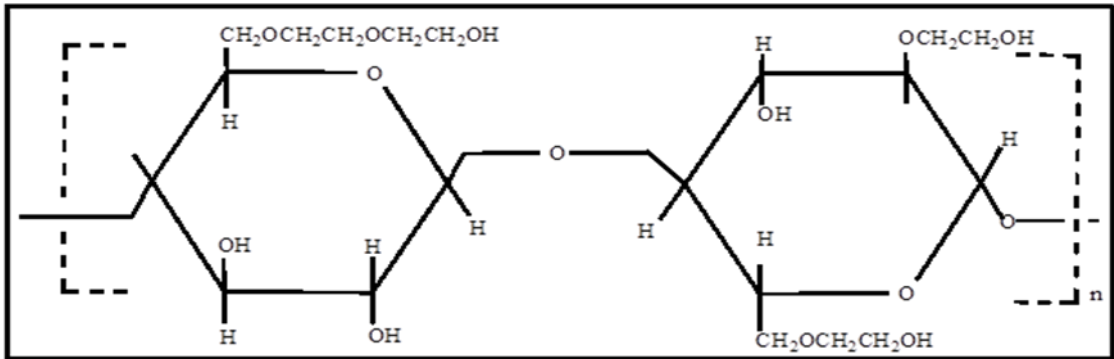


Fig 5 Molecular structure of hydroxyethyl cellulose [57].

2.4. Hydroxypropyl cellulose (HPC)

Sodium cellulose (Na-cellulose) is created when cellulose reacts with sodium hydroxide (NaOH). Hydroxypropyl cellulose (HPC) is produced by reacting sodium cellulose (or alkaline cellulose) with propylene oxide under high temperature and pressure. Propylene oxide with one or more of the three reactive hydroxyl groups present in each glucose monomer in the cellulose chain may create ether bonds under these circumstances. The degree of substitution (DS) varies in the range 1-3. However, if the second hydroxy group in the side chains reacts with propylene oxide, the DS value can be higher than 3. The obtained HPC is used in various industrial applications as colloidal stabiliser, flow regulator and surfactant. HPC exhibits heterogeneity because of the widely scattered arrangement of raw cellulose and the dispersion and allocation of propylene oxide throughout the polymer chain [58-61].

Hydroxypropyl cellulose is the most important cellulose derivative due to its easy production, complex phase behavior and hydrophilic properties [62]. Hydroxypropyl cellulose has a slightly yellowish white color and is a non-toxic, odorless and tasteless powder [48, 63]. As an application area, extruded films are widely used in food, pharmaceutical formulations and cosmetic products [48, 64, 65, 66]. It is used especially as a precipitant in many materials such as coating, encapsulation, bonding materials, foam carrier, protected colloids and foods, paper, ceramics and plastics [67]. Hydroxypropyl cellulose is a semi-crystalline polymer that is hydrophilic and soluble in non-ionic water and polar organic solvents and pH insensitive [24, 65]. In particular, when dissolved in water, they can be used as thickeners, binders, emulsifiers and stabilizers [65, 68]. It decomposes in water at temperatures below 40°C and in specific polar organic solvents such as methanol, ethanol, and isopropanol [59]. Figure 6 illustrates the molecular arrangement of hydroxypropyl cellulose.

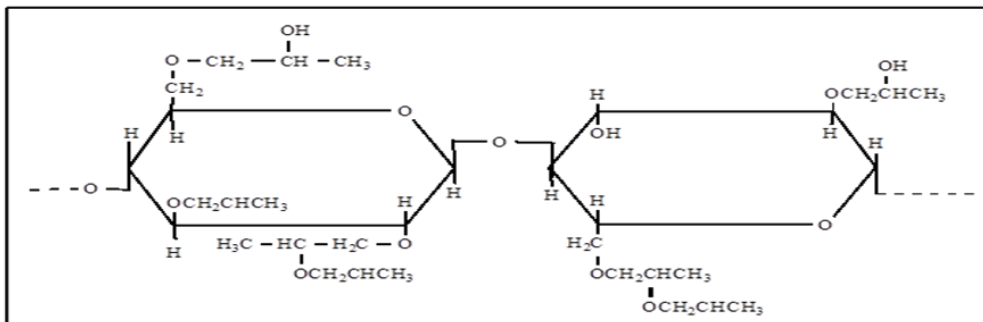


Fig 6. Molecular structure of hydroxypropyl cellulose [34].

2.5. Hydroxypropylmethyl cellulose (HPMC)

HPMC, a derivative of cellulose, is achieved through the reaction of alkaline cellulose with chloromethane and propylene oxide, resulting in partial o-methylation and o-(2-hydroxypropylation). HPMC is shown as the formula $C_8H_{15}O_8-(C_{10}H_{18}O_6)_n-C_8H_{15}O_8$ and its molecular weight is approximately 86000 [32, 69, 70, 71]. The term "molar substitution" (MS) in HPMC refers to the amount of hydroxypropyl groups each repeating unit, whereas "degree of substitution" (DS) refers to the number of methyl groups per unit [72-73.]

This cellulose derivative is a non-ionic polymer, soluble in cold water (below 40°C) but thermally reversible when heated in the range 50-80°C and turns into a relatively hard gel. It has a creamy or white hue, odourless, transparent, stable, oil-resistant and tasteless granular or fibrous powder. The solubility of HPMC, which has no toxic effect, is independent of pH (in the range of 1-10) and ionic strength, because there are no groups or permanent charges that can turn into ions in its structure. The lower the viscosity, the higher the solubility. At low temperatures, HPMC becomes more soluble in water, whereas at high temperatures, it becomes less soluble. This is due to the fact that water molecules and HPMC's hydroxypropyl groups may establish hydrogen bonds. The hydrogen bonds that hold water molecules together are more stiff on hydrophobic surfaces. The HPMC/water combination is transparent at low temperatures, and water molecules near hydrophobic methyl groups are arranged in a highly organized fashion. The system's kinetic energy grows with temperature, causing the hydrogen bonds to progressively weaken until the water molecules' lattice structures collapse and methyl groups emerge. At this point, the methyl groups' hydrophobic interactions start to encourage the development of a physical network with hydrophobic junctions. The gelation point of HPMC varies between 50 °C and 90 °C [69-74].

HPMC, among cellulose ethers, is commonly employed as the primary active component in hydrophilic matrix tablets designed for sustained release. At the same time, their roles as gelling agents hold significant importance in formulation procedures, as they contribute to the creation of a gel layer that resists diffusion and erosion through hydration, a crucial aspect in regulating drug release [43]. It also has a high swelling capacity, which significantly affects the release kinetics of the combined drug. It expands on contact with water or biological fluids and causes the polymer chains to relax, leading to an increase in its volume [75].

HPMC has several functions in the food industry as a film-forming, emulsifying, foaming, solubilising, stabilising, suspending and thickening agent. At the same time, HPMC serves as a semi-synthetic, viscoelastic polymer, inert and lacking in electrical charge, and is extensively employed as a lubricant in formulations related to ocular care, pharmaceuticals, medicine, food, and cosmetics [69, 74, 76, 77, 78]. Figure 7 depicts the molecular arrangement of hydroxypropylmethyl cellulose.

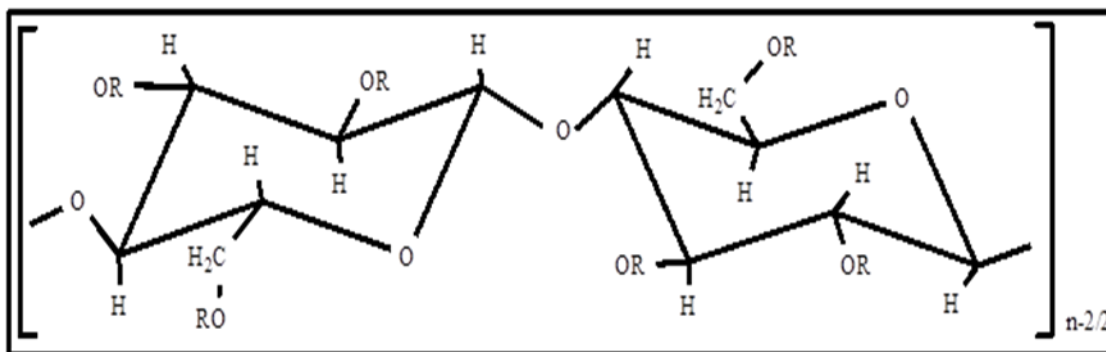


Fig 7. Molecular structure of hydroxypropyl methyl cellulose [79].

2.6. Carboxymethyl cellulose (CMC)

Carboxymethyl cellulose (CMC), a cellulose derivative, is a linear polysaccharide made up of anhydroglucose units that is both water-soluble and anionic. The repeating units are linked by β -1,4-glycosidic bonds. At the molecular scale, CMC and cellulose differ primarily in that CMC incorporates anionic carboxymethyl groups (CH_2COOH) within its structure, substituting certain hydroxyl groups found in pure cellulose. CMC was initially synthesized in 1918. According to the Williamson ether synthesis, the production of CMC involves two main stages: i) the alkaline cellulose process and ii) the esterification reaction. Initially, cellulose is treated with sodium hydroxide to produce active alkaline cellulose. This alkaline cellulose is then reacted with monochloroacetic acid. During this step, carboxymethyl groups replace the three hydroxyl groups in the cellulose molecules, with the degree of substitution ranging from 0.4 to 1.5. As a result of this substitution range, CMC can have molecular weights varying from 90,000 to 2,000,000 g/mol. However, the commercial production of these significant polymeric materials began in Germany in the early 1920s [80-81].

CMC is a crucial industrial polymer with diverse applications, such as entrainment reduction in sedimentation processes, in detergents, textiles, smart packaging industry, industrial construction and agricultural soil improvement, paper, food, pharmaceutical products, paint, pharmaceuticals and oil well drilling. It is also known as a viscosity increasing agent, widely used in pharmaceuticals, foods and cosmetics [21, 80, 82, 83, 84, 85] CMC forms through the interaction between alkaline cellulose and either monochloroacetic acid or Na-monochloroacetate via the Williamson ether synthesis process [12, 80, 86].

CMC stands as a paramount cellulose derivative, holding immense significance across industrial sectors and within our everyday routines. CMC, an anionic polysaccharide, exhibits water solubility and chemical reactivity owing to its elongated linear structure, featuring polar carboxyl groups derived from cellulose [11, 21, 87]. Moreover, refined cellulose exhibits a neutral flavor, lacks scent, and appears as a smooth ivory powder that flows freely. In addition to these properties, they have high molecular weight properties because they are water-soluble heteropolysaccharides. As a result, blending CMC with starch is frequently done to achieve the preferred consistency, enhance product excellence and durability, manage moisture levels, and guarantee optimal water movement [21, 87].

The characteristics of CMC change based on the level of substitution (DS, indicating the quantity of carboxymethyl groups in each anhydroglucose unit), the arrangement, and the polymerization extent. DS has the most significant influence on the solubility of the CMC solution, shear force, rheological characteristics, and resilience to temperature variations and the presence of low molecular weight additives [21]. The structural arrangement of carboxymethyl cellulose is depicted in figure 8.

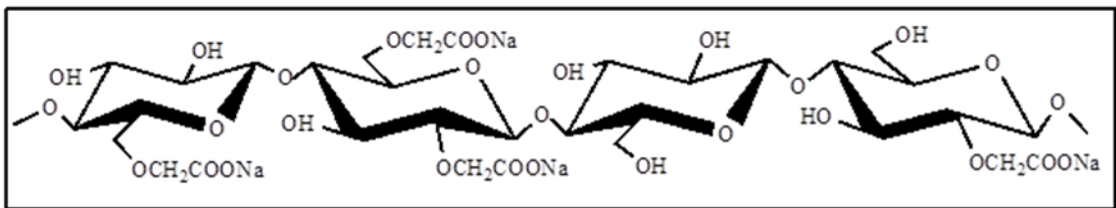


Fig 8. Molecular structure of carboxymethyl cellulose [87].

2.7. Sodium carboxymethyl cellulose (NaCMC)

NaCMC, a derivative of cellulose, which is the abundant polysaccharide composing the structural tissue of plants, is known as sodium carboxymethyl cellulose [88]. NaCMC is an anionic, linear cellulose derivative created through the reaction of cellulose with an alkali and chloroacetic acid. In this reaction, the H atom in the hydroxyl group of cellulose is replaced by the carboxymethyl group. As a result, the natural CH_2OH group in the glucose unit of cellulose

is easily replaced by the carboxymethyl group, thus obtaining sodium carboxymethylcellulose (NaCMC) [88-91]. This change is determined by the degree of displacement (DS). At the same time, the chain length of both DS and polymer determines the solubility, viscosity and gelation power of NaCMC [88].

NaCMC is used in many fields such as coatings, laminates, optical films and pharmaceuticals, as well as in the paper industry due to its high modulus, hardness, durability, thickening and stabilising properties. It is also one of the most important derivatives of cellulose, widely used in the food and cosmetics industries due to its moisturising, anti-caking, foaming, volumising, gelling and brightening properties. Nowadays, it is becoming increasingly popular as a soil conditioner [89]. Their high viscosity at low concentrations is the key characteristic that makes them valuable for these applications. Furthermore, the most important disadvantages of cellulose and derivative films are their mechanical properties, brittleness and low vapour permeability [24, 93, 94, 95, 96, 97] This polymer has become one of the most preferred cellulose derivatives due to its thickening, suspending, stabilizing, binding and film-forming properties [24, 98].

NaCMC is a derivative of cellulose in powder form, which is white to off-white in color, straight-chain, non-toxic, odorless, biodegradable and abundantly available, soluble in hot or cold water [23, 93, 94, 99, 100]. The molecular structure of NaCMC is illustrated in Figure 9.

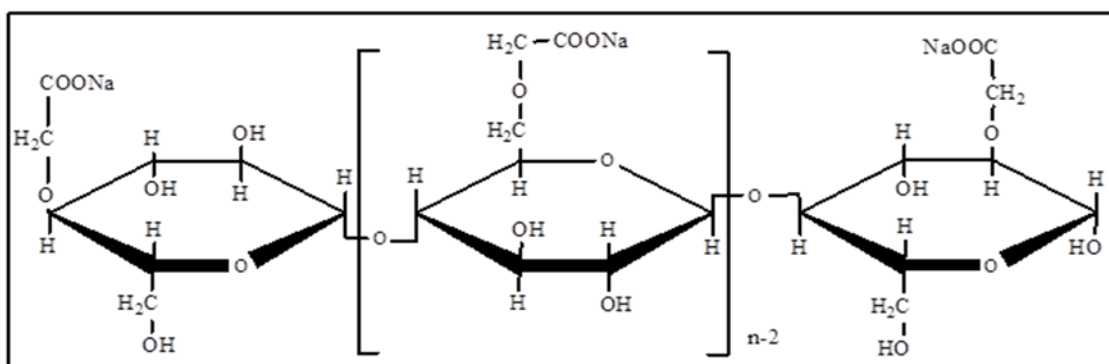


Fig 9. Molecular structure of sodium carboxymethyl cellulose [101].

The etherification and esterification derivatives of cellulose have many different uses and stand out as sustainable materials. These derivatives can show significant differences in their physical, chemical and mechanical properties depending on their environment and application conditions. The properties of various cellulose derivatives examined in detail within the scope of the study are presented comparatively in Table 1 and Table 2. In these tables, the properties of each cellulose derivative are summarized and their common applications are emphasized.

Table 1. Important properties of cellulose ethers considered in the study.

CELLULOSE ETHERS	PROPERTIES
Methylcellulose	<ul style="list-style-type: none"> ✓ pH value is between 2 and 12 ✓ Neutral, odourless, tasteless, inert and non-toxic and insoluble in water. ✓ Insoluble in most organic solvents. ✓ It is cost-effective and beneficial to the environment. ✓ It is synthesised by Williamson etherification method. ✓ It is used in pharmaceutical, chemical, ceramic, electrochemical supercapacitors and electrolyte applications. ✓ It gels in the range of 60-80 ° C temperatures. ✓ Polymerisation degree is in the range of 50-1000.

Ethyl cellulose	<ul style="list-style-type: none"> ✓ pH value is between 3 and 11. ✓ It is anionic and insensitive to pH. ✓ Insoluble in water. Dissolvable in organic solvents like alcohol, ether, ketone, and ester. ✓ Non-toxic and resistant to external factors. ✓ Its colour is white to light tan, odourless, tasteless powder and granular. ✓ It is used in areas such as optics, film formation, packaging, coating material and micro-encapsulation. ✓ Melting point is between 240 and 255 degrees Celsius. ✓ The transition temperature of glass is 120°C. ✓ Specific density is between 1.07-1.18, decomposition point 135-155°C and combustion temperature 330-360°C.
Hydroxyethyl cellulose	<ul style="list-style-type: none"> ✓ It is water soluble and has low density. ✓ Insoluble in most organic solvents. ✓ It is the most hydrophilic cellulose ether. ✓ It is an anionic substance with the ability to thicken, suspend, disperse, emulsify, create films, and protect against water.. ✓ It is used in polymer chemistry, ceramic industry, cosmetics, agriculture, construction materials, textiles, paper, paints, aerogel production, synthetic resins, etc.
Hydroxypropyl cellulose	<ul style="list-style-type: none"> ✓ It is anionic and semi-crystalline, soluble in most polar organic solvents and pH insensitive. ✓ It has a slightly yellow coloured white appearance. ✓ It is a non-toxic, odourless, tasteless powder. ✓ It is used in food, pharmaceuticals and cosmetics ✓ Used in industry as colloidal stabiliser, flow modifier and surface activator. ✓ It is used in coating, encapsulation, foam carrier, preserved colloid and food, paper, ceramics and plastics.
Hydroxypropylmethyl cellulose	<ul style="list-style-type: none"> ✓ Its solubility does not depend on pH value and ionic strength. ✓ It is a white or creamy white, odourless, transparent, stable, inert, oil resistant, non-toxic, anionic and tasteless granular or fibrous powder. ✓ Soluble in cold water below 40°C. ✓ It becomes a hard gel in the range of 50-80 °C. ✓ It is used in pharmaceutical, food and cosmetic fields. ✓ It has functions such as emulsifying, foaming, solubilising, stabilising, suspending and thickening agent.
Carboxymethyl cellulose	<ul style="list-style-type: none"> ✓ Anionic and soluble in water. Synthesised according to Williamson ether synthesis. ✓ It is a cream-coloured white, tasteless, odourless powder. ✓ It is used in detergents, textiles, smart packaging, construction, soil improvement, paper, food and paint.
Sodium carboxymethyl cellulose	<ul style="list-style-type: none"> ✓ It is odorless, non-toxic, soluble in both hot and cold water, and comes in powder form. It can decompose naturally. ✓ It is used in coatings, laminates, optical films, pharmaceutical and paper industry. ✓ It is widely used in food and cosmetics industry due to its moisturising, anti-caking, foaming, volumising, gelling and brightening properties.

3. Cellulose ester derivatives

Esterification, a common equilibrium reaction, involves the combination of an alcohol and an acid to produce an ester and water. Cellulose can undergo esterification with various acids, including acetic acid, nitric acid, sulfuric acid, and phosphoric acid [24]. Cellulose esters typically exhibit low solubility in water and possess excellent capabilities for forming films. Cellulose esters are widely used in pharmaceutical formulations for controlled release, such as in osmotic and enteric-coated drug delivery systems. These polymers are often used in combination with cellulose ethers to create microporous distribution membranes. Cellulose esters are divided into two distinct categories: one organic and the other inorganic. Cellulose esters derived from organic sources hold greater significance within the pharmaceutical sector. Different forms of organic cellulose esters find application in both commercial products and

pharmaceutical research. Organic cellulose esters like cellulose acetate (CA), cellulose acetate phthalate (CAP), cellulose acetate butyrate (CAB), cellulose acetate trimellitate (CAT), and hydroxypropylmethyl cellulose phthalate (CATM) exemplify this group. Organic cellulose esters hold greater significance in the pharmaceutical sector compared to their inorganic counterparts like cellulose nitrate and cellulose sulfate. Cellulose nitrate, also known as pyroxylin, possesses transparency and excellent film-forming properties. However, its infrequent utilization in pharmaceutical formulations stems from its minimal flammability in contemporary pharmaceutical solvents [19].

The presence of three hydroxyl (OH) groups in each glucose unit allows for the formation of mono-, di-, and tri-ester compounds. The mutual bonding of hydrogen bonds and OH- groups within the large molecular structure of cellulose causes this structure to be partially or completely broken during esterification. As the cellulose groups attack the cellulose chain in pieces, the chain structure of the cellulose either transforms into another structure or degradation occurs [12]. The esterification reaction of cellulose is shown in figure 10.

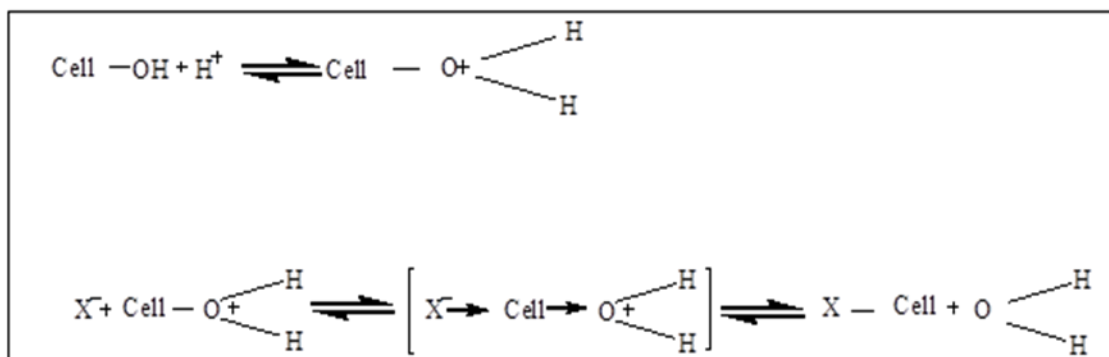


Fig 10. Esterification reaction of cellulose [12].

3.1. Cellulose acetate (CA)

Cellulose acetate (CA) is a widely used derivative of cellulose, the main component of plant cell walls. It can be produced from diverse natural resources including cotton, recycled newsprint, sugarcane pulp and straw, rice husk, sorghum straw, and Babassu coconut shells [102-103].

Cellulose acetate (CA) is a significant cellulose ester produced by reacting cellulose with acetic anhydride and acetic acid in the presence of sulfuric acid. This process facilitates the replacement of cellulose's hydroxyl groups with acetyl groups [23, 102, 104]. The degree of substitution (DS) significantly influences the material's properties, including its thermal behavior, hydrophobicity, transparency, processability, and solubility. Notably, hydrophobicity and processability distinguish cellulose acetate (CA) from natural cellulose, which is insoluble in most solvents. Additionally, as the DS increases, the glass transition temperature of cellulose acetate generally decreases [105]. The molecular structure of cellulose acetate is given in figure 11.

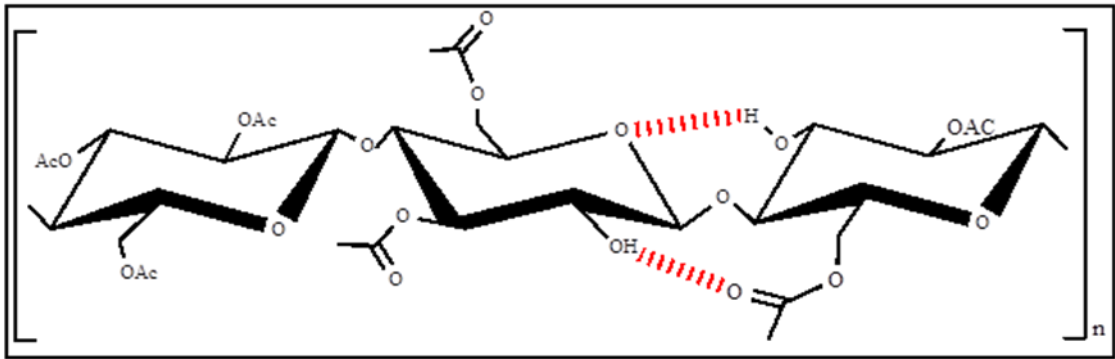


Figure 11. Chemical structure of cellulose acetate [106].

Cellulose acetate stands out as the most important biopolymer cellulose derivative obtained from organic acids. Thanks to its superior physical, chemical and biological properties, it finds wide application in many fields; varnishing, textiles, photographic films, active food packaging, removal of pollutants from waste water, plastic materials, membranes, cigarette filters, medical applications, film and fibre production [12, 105, 107, 108, 109, 110, 111, 112]. Compared to cellulose nitrate, its flammability and combustion properties are quite low. At the same time, film strips made of cellulose acetate can remain intact for many years [12]. CA is a biocompatible, biodegradable, and non-toxic biomaterial. It is relatively inexpensive, has low hygroscopicity, and exhibits excellent resistance to chemicals and heat [102, 113, 114]. Application areas that benefit from cellulose acetate are given in figure 12.

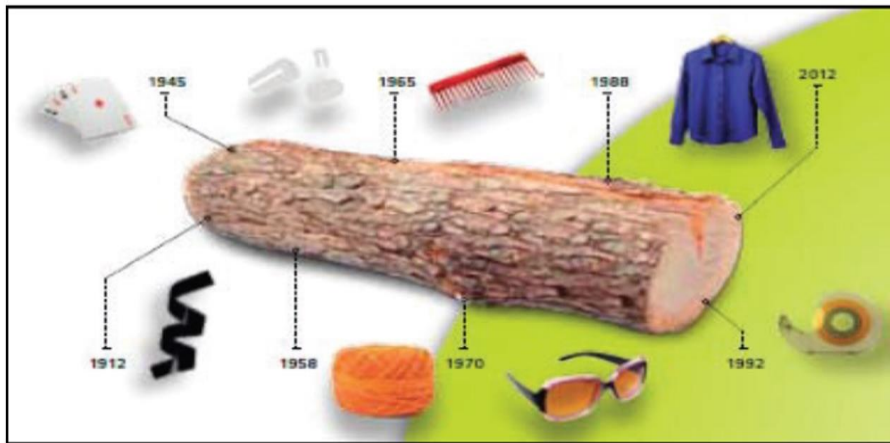


Fig 12. Application areas that benefit from cellulose acetate from past to present [106].

The mechanical strength, superior abrasion resistance, transparency, heat resistance, low water absorption, dyeability, simplicity of processing, moldability, and high dielectric strength of cellulose acetate are among its noteworthy qualities [12, 111, 121]. Cellulose ester derivatives like cellulose acetate-butyrate and cellulose acetate propionate hold more significance compared to standard cellulose acetate owing to their attributes, including minimal moisture absorption, heightened dimensional stability, exceptional resistance to atmospheric conditions, superior abrasion resistance, dyeing capabilities, and enhanced finishing techniques [12].

Technical properties determined by the degree of substitution are very important for cellulose acetate in terms of determining its solubility in organic solvents, its behavior in areas of use such as plastic material production and varnish resin production. At the same time, the structure responsible for the mechanical and processability properties of cellulose is called viscosity, that is, the degree of polymerization [12].

3.2. Cellulose nitrate (NC)

Cellulose nitrate (cotton powder) is an important cellulose derivative, a semi-synthetic plastic produced by the reaction of cellulose with inorganic acids. This polymer, produced through the nitration process involving a mixture of nitric acid, sulfuric acid, and water, is made up of d-glucopyranose units connected by β (1-4) glucosidic bonds. As a cellulose derivative, its structural distinction comes from the substitution of hydroxyl groups (-OH) in cellulose with nitrate groups (-ONO₂) [12, 115, 116]. Cellulose nitrate is used in various fields depending on its nitrogen content. These include plastics (celluloid), varnishes, energy binders, rocket propellants, smokeless gunpowder, civil and military applications, paints, production of new filter membranes, composite coatings, adhesives and explosive making. In particular, cellulose nitrate with a nitrogen content of less than 12% is used in the production of photographic films, inks and paints, while those with a nitrogen content of more than 12% are used as propellants in explosive formulations [12, 117, 118, 119, 120].

The degree of substitution of NC can be determined depending on the degree of polymerisation (DP) or nitrogen content of the cellulose. Differences in this degree of substitution also lead to variations in the products obtained. Furthermore, the different rates of substitution of cellulose nitrate also affect its solubility in organic solvents. [12]. Cellulose acetate (NC) is considered one of the most appealing and promising energetic materials for both current and future uses, thanks to its remarkable characteristics, including low cost, high flammability, explosive potential, and high enthalpy of combustion. [120]. However, cellulose nitrate poses a serious risk to museum and archive collections. This cellulose derivative can degrade substantially over time, leading to yellowing, brittleness, and cracking, which may eventually cause the material to disintegrate completely. It can also be dangerous as it releases nitric acid, a strong oxidiser and corrosive in its environment [119].

In addition to the previously mentioned applications of cellulose nitrate, it is also valued for its thermoplastic properties. Its ability to melt easily when heated makes it different from rubber. Another distinguishing characteristic of cellulose nitrate, compared to rubber, is its elastomeric property. Thanks to its elastomer feature, it prevents the shape from deteriorating easily, thus preserving the shape. While the nitration process for commercial purposes is carried out using sulfuric acid and aqueous nitric acid, the raw material of cellulose is obtained chemically from wood or pure cotton or cotton threads. To prevent issues like delignification and degradation caused by alkaline extraction during the cellulose extraction process and to achieve high degree of polymerization (DP) values, cellulose is sourced from wood that has undergone a nitration process. As a result of this application, the polymerization degree (DP) of different wood types was found to be between 5000 - 10,000. In studies conducted on this subject, it has been determined that mixtures rich in HNO₃ have a low degradation effect on cellulose. It has been observed that treating cellulose for a long time and the high temperature effect increases the yield obtained from extraction [112].

Table 2. Important properties of cellulose esters considered in the study.

CELLULOSE ESTERS	PROPERTIES
Cellulose acetate	✓ Used in varnishing, textiles, photographic films, active food packaging, waste water cleaning, plastics, membranes, cigarette filters, medical applications, film and fibers.
	✓ It has low ignition and combustion properties.
	✓ Non-toxic, low cost, chemical and heat resistant.
	✓ Mechanical durability, high abrasion resistance, low water absorption, dyeability and processability.

Cellulose nitrate	✓	It is a semi-synthetic plastic.
	✓	It is used in plastics, varnishes, rocket propellant formulations, smokeless gunpowder, civil and military applications, paints, composite coatings, adhesives and explosives.
	✓	Its low cost, high flammability and explosive properties make it attractive for future applications.
	✓	It is not recommended for use in museum and archive applications as it will deteriorate over time, causing yellowing, brittleness and cracking.
	✓	It is dangerous in environments where it releases nitric acid.
	✓	The degree of polymerization is between 5000-10000.

4. Conclusion

In the 1970s, with the industrial revolution, steam engines began to be used, and this caused a simultaneous increase in the use of synthetic products. The use of these unnatural products has started to have negative effects on the environment over time. Among these negative effects, the fact that the materials used remain intact in nature for many years after their useful life has ended has begun to be seen as the most important problem. As a result of these developments, searches have begun to use renewable and clean raw materials. One of the results of these searches is the use of cellulose raw material. Cellulose derivatives produced from cellulose raw material can also be considered in this context. It seems that they have significant advantages compared to petrochemical products, especially in terms of their rapid decomposition in nature after their use. At the same time, Cellulose derivatives, created through the modification of cellulose, possess robust, cost-effective, reproducible, recyclable, and environmentally friendly qualities.

Conflict Declaration

As the authors of this study, we declare that we have no declaration of conflict.

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