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ARTICLE

Cellulose at Scale: From Past Challenges to Future Opportunities for Sustainable Functional Materials

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Cellulose-based chemistry was at the origin of modern materials science and has today re-emerged as one of the most dynamic and rapidly evolving research fields. How did a biopolymer that enabled early industrial breakthroughs such as celluloid follow such an uneven scientific trajectory, and is it now re-emerging as a platform for future materials? In this Perspective, we revisit the non-linear evolution of cellulose chemistry to identify the scientific, technological, and conceptual barriers that have long constrained its development, despite its abundance and renewability. We argue that cellulose must play a pivotal role in future materials and chemical industries within a sustainable technological ecosystem, and we outline the chemical, structural, and analytical challenges, and the corresponding design principles needed to fully unlock the potential of this inherently complex polysaccharide.

Cellulose research encompasses a vast landscape, with numerous reviews covering; nanotechnology, packaging, composites, biomedical technologies, fiber science, and energy systems. Scientific approaches and challenges to these fields are equally diverse: for instance, the physics of colloidal nanoscale cellulose,[1] the engineering challenges of fiber processing,[2] or the biological studies linking biosynthesis mechanisms to cellulose.[3] In this perspective, we step back and examine the rich, complex history of cellulose science, through the lens of chemistry towards sustainable functional materials. By understanding our longstanding relationship with this biopolymer - why it was historically utilized, in what contexts, and why its relevance has evolved - we hope to show the importance of cellulose as a biopolymer platform in our collective transition toward a more benign materials economy, while also recognizing the inherent challenges this also presents.

To this end, our focus is deliberately narrower: rather than addressing cellulose science in its entirety, we center on its chemistry, specifically post-polymerization modification of naturally produced cellulose from the perspective of polymer science, excluding areas such as degradation into platform chemicals or colloidal applications. Valorizing cellulose at the macromolecular level offers substantial sustainability benefits, as it leverages nature's pre-formed structures. However, this also requires us to adapt our material vision to its unique polymeric features, work with, rather than against its intrinsic properties, limitations, and specific requirements.

The Rich History of Cellulose

From Nature to the Service of Humanity

To begin with, it is important to consider what cellulose is, how it was first discovered, as its historical context is important for understanding both its significance and its future potential. Fundamentally, cellulose is a polysaccharide composed of repeating $\beta_{1\rightarrow4}$ linked D-glucose units. Cellulose, found in most plants and trees as well as in certain bacteria, fungi or algae, it constitutes roughly 25 - 40 % of the total global biomass, equivalent to approximately 300 to 500 Gt dry mass .[4, 5] It represents around 30 - 50% of plant biomass, which itself accounts for about 80% of the estimated

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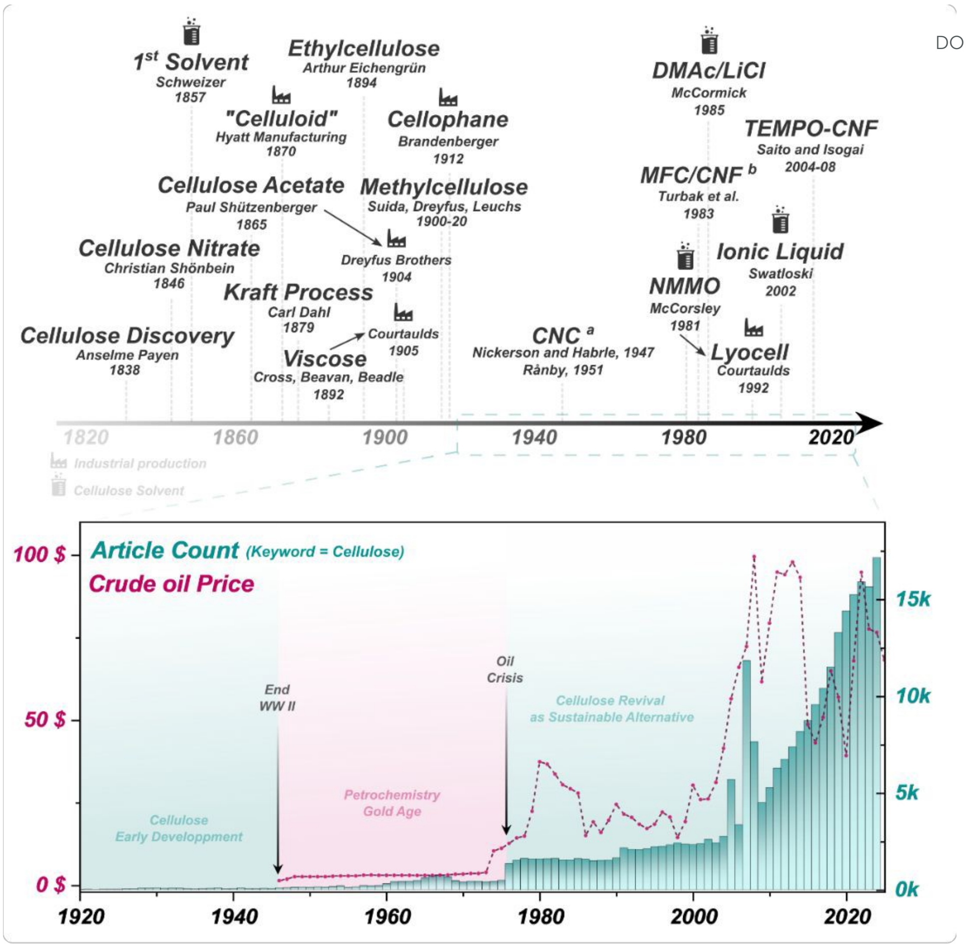

Figure 1. Top: Key dates and milestones in cellulose research and technology (^{a}CNC refers to cellulose nanocrystal; ^{b}MFC and CNF refer respectively to microfibrilated cellulose and cellulose nanofibrils). Bottom: Historical trends in crude oil prices (source: Macrotrends) and the number of scientific publications related to cellulose (source: Web of Science – Clarivate).[6] A partial correlation can be observed between oil prices and scientific interest in cellulose, even when accounting for inflation effects (oil price and scientific production). The post-World War II era marked the advent of petrochemistry; 1973 and the first oil crisis triggered renewed interest in cellulose, driven by emerging sustainability concerns.

550 Gt C of total biomass on Earth. [7, 8] Cellulose predominance in living systems can be attributed to its unique properties.[9] Strong and stable cellulose-cellulose interactions give rise to highly crystalline (60 to 80%),[7, 8] rigid and robust structures, making it for instance an ideal building block for load-bearing applications.[10, 11] Furthermore, cellulose biosynthesis is both efficient and energetically favorable for plants: starting from abundant glucose, the polymer is synthesized and extruded through the plasma membrane by cellulose synthase complexes. [7, 8]

Humankind has been relying on cellulose it since early times, employing this material for textiles (e.g., linen), as writing media: from papyrus in ancient Egypt (~ 3000 BC) to the first reported paper by Ts'ai Lun in China around AD 105,[12, 13] but also in polymer science. In fact, the first molecular structure deciphering of cellulose through elemental analysis dates back all the way to 1838 by the French scientist Anselme Payen (further complemented by Staudinger polymeric understanding in 1920), marking the premodern dawn of a new era in materials chemistry and science.[14, 15]

Pioneering of the Plastics Age

Following Payen's discovery, cellulose knowledge and application space developed alongside synthetic chemistry, as illustrated in Figure 1. The first cellulose derivative, nitrocellulose (cellulose nitrate), was developed in 1846 by Christian Shönbein through the nitration of cotton with nitric acid and sulfuric acid, [16-18] followed by its first industrial production as "Celluloid" in 1870 (Hyatt Manufacturing Company) which had a profound impact on both chemistry and manufacturing.[19] As one of the first synthetic thermoplastics, Celluloid represents an important contribution to early polymer science by providing a new class of moldable materials that could be produced on an industrial scale. Other landmark derivatives followed. In 1892, the viscose process was developed,[20, 21] involving the derivatization of cellulose with carbon disulfide under alkaline conditions, followed by

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hydrolysis and regeneration. This innovation was a major step in the industrial use of cellulose, enabling the production of continuous filaments for rayon fibers and large-area transparent films. Rayon, produced via the viscose route, became the first commercially successful man-made fiber, providing a cheaper and more versatile alternative to silk and natural fibers in textiles. At the same time, this process led to the creation of cellophane, a transparent film of regenerated viscose invented and commercialized by Brandenberger in 1912.[22] Cellophane revolutionized packaging: its transparency, flexibility, and relative low cost made it ideal for wrapping food, confectionery, and consumer goods, thereby changing retail display and consumer habits. In technical terms, the viscose process also demonstrated that cellulose could be dissolved, reshaped, and regenerated on a large scale.

Cellulose acetate is another pivotal derivative, whose development started as early as 1865 with the work of Schützenberger and reached industrial scale in 1904. [23, 24] Today, it remains one of the most widely produced cellulose thermoplastics, with an estimated global production around 1-2 Mt in 2024. [25-28] This sustained output reflects cellulose acetate unique position as bio-based, processable thermoplastic combining good optical clarity and dyeability in demanding yet niche applications (Table 1). The family of cellulose ethers also emerged as highly versatile derivatives, valued for their solubility in water or common organic solvents, enabling their widespread use as functional additives (Table 1).[29] Key examples include carboxymethylcellulose (1918),[30] hydroxyethyl cellulose (1920),[31-33] methylcellulose (1900–1920), [34-37] or ethylcellulose (1894). [29] Together, cellulose esters and ethers exemplify how chemical derivatization transformed an insoluble biopolymer into a platform of processable, functional materials. In this sense, cellulose was a vital raw material for the society and for the industrial transformations of the 19th century, standing as a central driver of the first polymer-based materials economy.[16] Yet, this central role gradually diminished in the decades that followed, as the post-WWII period marked the golden age of petrochemistry, during which industrial and academic research increasingly focused on this versatile, and economically competitive resource.[38] Consequently, cellulose gradually lost popularity due to cost competitiveness, despite significant advances during this time, such as the first descriptions and production of cellulose nanocrystals (CNC) which would only gain widespread attention decades later. [39, 40] Commodity plastics and resins, like polyolefins, nylons, and polyurethanes, rapidly outperformed cellulose derivatives in terms of properties, cost and processing efficiency.[38] Thus, rather than disappearing, cellulose chemistry entered a phase of relative dormancy, overshadowed by the rapid expansion of fossil-based polymer technologies.

A Key Alternative for a Sustainable Material Economy

As the world began to question the production, use, and environmental impact of petroleum hydrocarbons, particularly after the price surge during the first oil crisis in 1973 (Figure 1), a slow but steady shift in public perspective emerged. [41] Growing awareness of global warming and sustainability challenges has since then gradually reshaped our priorities. [42, 43] The search for alternative renewable and bio-based raw materials is becoming increasingly urgent in a material economy still overwhelmingly dependent on fossil resources. To give an idea, around 92 % of all polymers produced today are fossil-based, including 99 % of structural polymers as well as around 60 % of all fibers. [28, 44, 45] These figures illustrate both the scale of the challenge and the need for new abundant and renewable polymer platforms.

In this context, cellulose has reemerged as one of the most promising renewable carbon resources, occupying many technologically relevant niches, as summarized in Table 1 for the most representative ones. Nonetheless, except for fibers and cellulose esters, cellulose derivatives are rarely employed as standalone materials or as the primary structural component in polymer systems. This limited use reflects not only economic and processing constraints, but also fundamental challenges related to synthesis, substitution control, structure–property relationships, and scalability. It is not a surprise that industrial scale derivatives of celluloses still rely on relatively old efficient synthetic routes (Williamson reaction, anhydrides, alkyl halides). As previously discussed, the relatively narrow use of cellulose derivatives should not be regarded as a limitation but as an opportunity, a challenge that invites innovation. Further valorization of cellulose is both desirable and essential, especially given the scale of the challenge: global plastic production now exceeds around 400 Mt per year. [28, 46, 47] Reducing the dependence on fossil-based resources requires alternative feedstocks of comparable or greater magnitude, among which cellulose represents a particularly promising candidate.

Cellulose Abundance at Scale

This perspective raises naturally the question of how much cellulose is actually available, and in which forms. Global assessments of plant biomass (450 Gt C) and net primary production (NPP, 60 Gt C/y), combined with the assumption that carbon constitutes approximately 45 % of dry plant biomass (of which around 30–50 % is cellulose) enable an estimation of the global cellulose pool. Based on these parameters, the total standing stock of cellulose is in the range of 300 to 500 Gt (dry mass), with an annual bioproduction or renewal rate of 40 to 65 Gt (Figure 2). [48, 49] At the beginning of the 21st century, human activities were estimated to appropriate approximately 15 Gt C of terrestrial biomass per year, corresponding to ca. 10-15 Gt/y of cellulose dry mass, predominantly derived from forestry and agricultural production.

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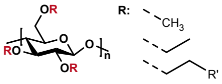
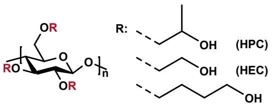
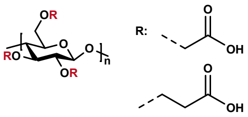
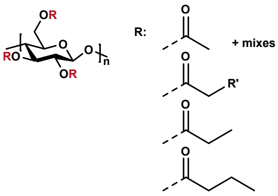
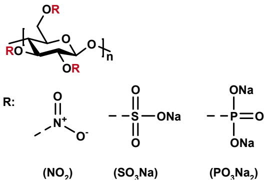
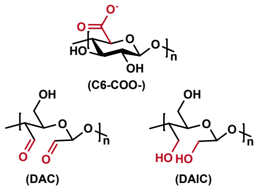
	Synthesis	DS	Properties	Applications	Refs
Alkyl Ethers 	Williamson: Alkali treatment → Alkyl halide	1.5-2.5	DS driven solubility (water, organic solvent), amorphous, T_g 130-200 °C, reversible gelation (MC)	Additives (Food, Pharma, Construction, Cosmetics), Coatings	9, 29, 34-37, 50, 51
Hydroxyalkyl Ethers 	Williamson: Alkali treatment → Hydroxyalkyl halide / Epoxide	**MS** **(molar subs.)** 1.5-4.0	**HPC**: reversible gelation, Tg< RT, T_{deg} 300°C **HEC**: Solubility (Water, polar solvent), Tg 120-150 °C, T_{deg} 200 °C	Coatings & Additive (Thickener, Binder, excipient). Construction, Pharma, Drilling	9, 31-33, 50-52
Carboxylated Ethers 	Williamson: Alkali treatment → chloroacetate	Mainly 0.4-1.2 (≤ 1.5)	Solubility (water), charged polymer (pH dependent)	Additive & Coating, Polyelectrolyte application, Nano-objects & colloids	30, 50, 51, 53-55
Esters 	Main Route: Anhydride, acid cat., Anhydride/acid/mix as solvent Secondarily: via Acyl chloride in organic solvent	Mainly 1.5 – 2.5	DS driven solubility (polar organic solvent), Tg 120 – 200 °C, thermoplastic behavior, mostly amorphous, transparent & low haze. Hydrolysis sensitive	Fibers, Films, Membranes, Plastic, Coatings	9, 50, 56
Heteroatom Esters 	**NO_2**: Nitration (HNO_3/H_2SO_4) **SO_3Na**: Sulfonation (SO_3, $ClSO_3H$) + neutralization **PO_3Na_2**: Phosphorylation (H_3PO_4 + urea/$POCl_3$) + neutralization	**NO_2**: 1.0-3.0 **SO_3Na**: ≤1.0 **PO_3Na_2**: ≤1.0	**NO_2**: Solubility (acetone, alcohol), flammable and explosive **SO_3Na**: Solubility (water), polyelectrolyte behavior **PO_3Na_2**: Solubility (water), polyelectrolyte behavior, chelating	**NO_2**: propellant, explosive, Coatings **SO_3Na**: Colloids, Hydrogels, Biomedical **PO_3Na_2**: Biomedical, Colloids, water treatment	16-18, 50, 57
Oxidized Cellulose 	**DAC**: $NaIO_4$, water **DAlC**: DAC reduction ($NaBH_4$) **C_6-COO-**: TEMPO / NaBr / NaClO oxidation	**DAC, DAlc** oxidation ≤100% **C_6-COO-**: 0.1-0.5	**DAC**: -CHO functions, highly reactive, crosslinkable **DAlC**: hydrophilic, flexible, Tunable Tg 20-110 °C, thermoplastic **C_6-COO-**: Polyelectrolyte, high dispersibility and stability	**DAC**: Hydrogel, biomedical, paper **DAlC**: Coatings, films **C_6-COO-**: Nano-objects, Rheology, Films, Hydrogels	58-63

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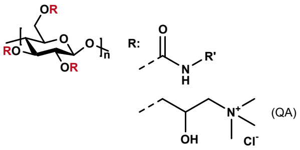
N-Based Derivatives 	**Carbamates**: R'=H urea/NaOH. R'≠H isocyanates **QA**, Williamson: Alkali treatment → Halide / Epoxide aqueous cond.	**Carbamates**: 0.1-1.0 **QA**: 0.05-0.4	**Carbamates**: R'=H alkali soluble and reactive, H-bonding, tunable polarity, thermoplastic **QA**: Cationic polyelectrolyte	**Carbamates**: Fibers, Membranes, Chromatography **QA**: Colloids, Additive, Rheology	[50, 56, 64, 65]

Table 1: Representative Cellulose Derivatives and Their Distinct Property Windows, Main synthetic pathway and application

Figure 2: Relative magnitude of global cellulose resources (stock, harvested, and accessible fractions) and plastic production in 2025. Cellulose stock and usage is estimated on the total carbon bioproduced and harvested [27, 48, 49, 66] Oil extraction and plastic production values show the comparative scale of the challenge of our fossil-based material economy. [28, 46, 67]

In terms of biogenic carbon, few other candidates can compete. Lignin represents a complementary counterpart to cellulose but requires separate considerations.[68-70] Another linear polysaccharide, chitin/chitosan, is bioproduced in substantial quantities (≤ 1 Gt/y), yet it remains far less abundant and its commercial production is limited to ∼100 kt/y.[71-74] These considerations highlight the importance of cellulose for scalable sustainable material production.

However, realizing the potential of cellulose at scale requires that its extraction and utilization is carried out wisely and in a sustainable way.[48, 75, 76] In this context, increasing biomass harvesting is feasible but must not exacerbate deforestation, biodiversity loss, or broader ecological stress. Fortunately, existing forestry infrastructure can facilitate this transition: the global forestry sector already produces around 150 Mt of wood pulp annually, of which approximately 2 % is high-purity dissolving pulp (> 93 % cellulose).[27] While the majority of this output is still devoted to paper products,[77] declining demand is accelerating efforts to develop advanced, high-value applications for cellulose.[78, 79] In parallel with primary streams, circular strategies such as recycling, reuse, and valorization of biomass side streams must complement these efforts. For example, the forestry, agricultural, and food industries generate together an additional 1.0–1.2 Gt/y of cellulose-rich waste, representing a valuable secondary resource for sustainable utilization (Figure 2). [48, 66, 80] The key therefore, is to bridge the gap between abundance of raw biomass with high-quality cellulose feedstocks that, in combination with advanced functionalization and processing, add further value to cellulose. Over the past decades, efforts to overcome this has triggered a new wave of technological innovation in cellulose fractionation, dissolution, and structure control, laying the groundwork for modern cellulose valorization.[81]

Modern Advances in Cellulose Valorisation

Building on technological advances since the 1970s, the valorization of cellulose has advanced significantly (Figure 1). For example, the 1980s witnessed the emergence of cellulose nanotechnology through the development of microfibrillated cellulose (MFC) and cellulose nanofibrils (CNFs), building on earlier work on cellulose nanocrystals (CNCs). These nanoscale building blocks opened new possibilities for the controlled design of advanced functional materials. [82-84] A landmark contribution in this context is the work of Saito and Isogai on the controlled oxidation of cellulose to obtain TEMPO-CNF, which established a mild, selective, and scalable route to highly dispersible cellulose nanofibrils with precise tunable surface chemistry.[59-61, 85, 86] In parallel, cellulose chemistry saw the development of powerful new solvents tailored to dissolve cellulose, enabling efficient homogeneous chemical modification, enabling both more detailed structural analysis, and advanced processing routes, like fiber spinning. [87-90] To cite a few, milestones like the introduction of DMAc/LiCl (1981),[91] NMMO (1981),[92-96] which led to the industrial-scale Lyocell process (1992),[97] or the first ionic liquids reported to dissolve cellulose by Swatloski et al. in 2002 collectively paved the way for a new directions in cellulose research. [90, 98] By allowing non-derivatizing dissolution of native cellulose without prior activation, ionic-liquid-based systems opened homogeneous routes to derivatization and processing that were difficult or impossible with earlier solvent platforms. Their broader deployment is nonetheless still constrained by cost, difficult solvent recovery and toxicity

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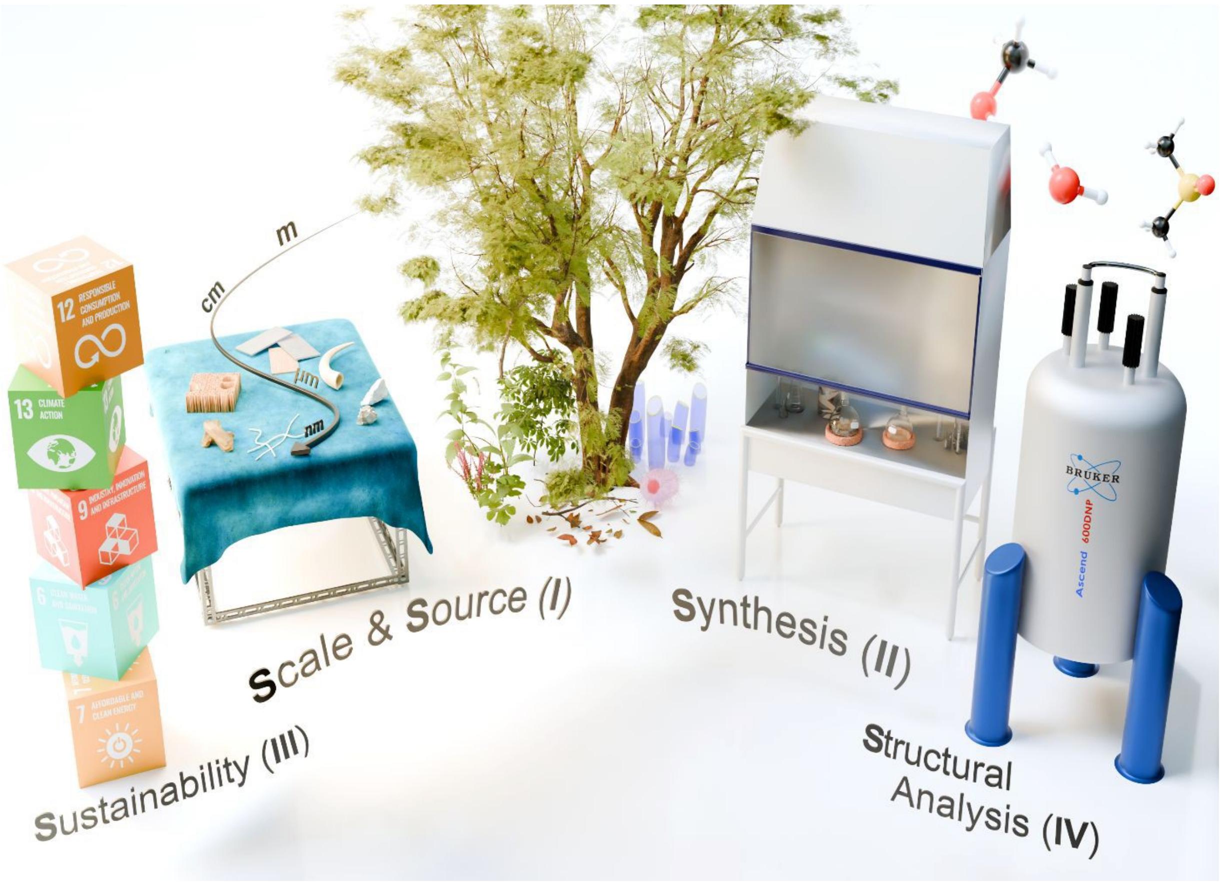

Figure 3: Cellulose chemistry's **5S** challenges (**S**ource, **S**cale, **S**ynthesis, **S**ustainability and **S**tructural Analysis)

management. Still, the Lyocell process introduced a largely closed-loop technology that reduce emissions and other environmental burdens compared with the viscose process while enhancing fiber performance, albeit at the cost of handling thermally unstable and potentially hazardous NMMO.

Despite this renewed momentum and regain in popularity for cellulose research over the last 50 years, there is still a clear reason why fossil-based synthetic materials were and remain king nowadays. [38] Materials like common polyolefins, PS, PET, Nylon, PVC are highly optimized, versatile, efficient and economically attractive. Comparably, working with cellulose still poses significant technological challenges, from its sourcing and processing to its chemical tuning and characterization.

Challenges in Using Cellulose as a Raw Material

Cellulose Beyond the Standard Polymer Paradigm

Although highly attractive as a polymeric raw material, cellulose is fundamentally different and does not conform to classical processing routes that underpin most commodity polymers. To exemplify, most common polymers are readily obtained by straightforward polymerization of their respective monomers. Direct polymerization from the monomer brings a number of inherent advantages and unlocks a wide range of possibilities offered by polymer chemistry, such as controlling the polymerization mechanism, incorporating co-monomers, tuning tacticity, stereochemistry or architecture.[99] Such control is largely inaccessible for cellulose, although laboratory-scale syntheses are reported in literature, for instance, via polycondensation, [100, 101] enzymatic polymerization, [102, 103] or

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through cationic ring-opening polymerization [104, 105] (notably for L-cellulose !). These methods, tedious to implement, remain intended to extend fundamental understanding or for niche and specialized high-value applications. In the current state, these results do not represent viable large-scale routes to produce cellulose or its derivatives (at least for now). [9, 106] Consequently, it is more advantageous to harvest and utilize cellulose as it is naturally bioproduced, which leads to the second limitation: cellulose degrades before melting or softening by reaching its glass transition (estimated between 220 and 250°C),[107, 108] which prevents melt processing without chemical modification.[109, 110] This contrasts with common thermoplastics that can be readily processed in extruders, film blown or injection-molded. This apparent disadvantage also highlights Nature's remarkable design: cellulose's unique supramolecular structure endows it with exceptional mechanical properties, including a theoretical Young's modulus of up to 143 GPa for the cellulose I$_\beta$ crystal, comparable to Kevlar's modulus.[111-113] Hence, from a chemical perspective, cellulose differs fundamentally from most conventional polymers. Realizing its full potential across a broad range of applications requires a thorough understanding of the unique challenges associated with its molecular and hierarchical structure. Here, we highlight these challenges around the five S's: Source, Scale, Synthesis, Sustainability, and Structural Analysis (Figure 3).

Scale and Source (I)

Cellulose complexity becomes even more evident when one considers that it is not a uniform material, but rather exists in a wide variety of assembled forms, which poses both a remarkable opportunity and an inherent challenge for its study and utilization.[4] Its purity, degree of polymerization (Figure 4), morphology or crystalline structure depend strongly on the source, the scale at which it is obtained, and the processing history it has undergone. [9, 114] As a result, each modification must be considered in tandem with the specific form of cellulose it is applied to. For instance, cellulose can be extracted from wood, which itself comprises a wide range of species (~80 000),[115] including softwoods and hardwoods.[116, 117] It can also be obtained from other plant fibers, including cotton, which offers high cellulose purity and DP, as well as flax, ramie, hemp or bamboo. [114, 118-120] Other more exotic sources also yield cellulose with incredibly high degrees of polymerization, crystallinity, or purity, such as bacteria or marine tunicates. [121, 122] For instance, tunicate cellulose is valued in fundamental research for its highly crystalline microfibrils with unusually large crystalline domains, making it ideal for structural elucidations (e.g., via AFM or scattering techniques) or for specialized high-value applications. [122, 127-129] Bacterial cellulose, on the other hand, is produced by unicellular microorganisms with remarkable purity, free from accompanying polysaccharides such as hemicelluloses found in wood. It presents high degree of polymerization values up to ~16 000 (~2600 kg/mol) with typical values ranging from 4000 to 10000. [121, 123, 124] These DP are comparable to, and in some cases higher than, those of many high-performance synthetic polymers, which is particularly striking given that cellulose is assembled biosynthetically via stepwise enzymatic chain growth (a step-growth mechanism) rather than via conventional chain-growth polymerization (Figure 4). Beyond these primary sources, a wide range of inexpensive and readily available secondary sources can also be valorized. These include forestry and agricultural residues (e.g., wheat and rice straw and husks, sugarcane bagasse, banana peels) as well as post-consumer textile and construction waste. [4, 114] Such residues offers large promising feedstocks (1.0 to 1.2 Gt/y, Figure 2) that could be upgraded into high-value cellulose materials rather than being simply burnt for energy. [130]

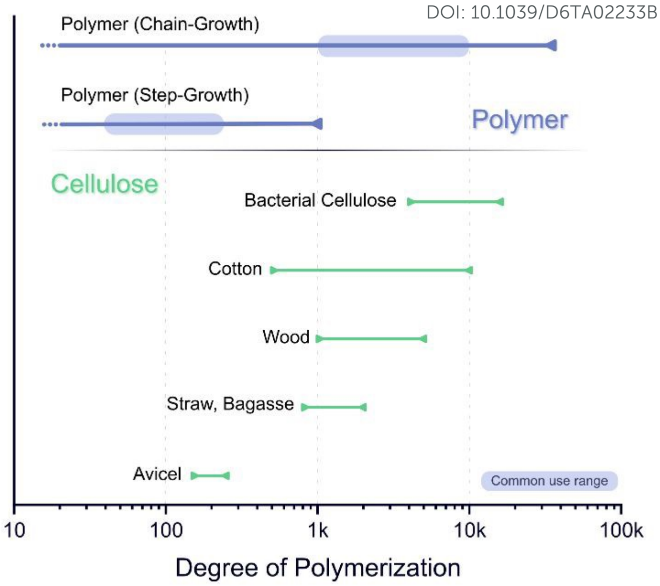

Figure 4: Degree of polymerization (DP) comparison between Cellulose sources and common synthetic polymers (obtained via chain-growth or step-growth mechanisms). Data reported in the literature for the DP of bacterial cellulose, [121, 123, 124] Avicel, [125, 126] and the other sources, [124] were primarily determined via viscometry.

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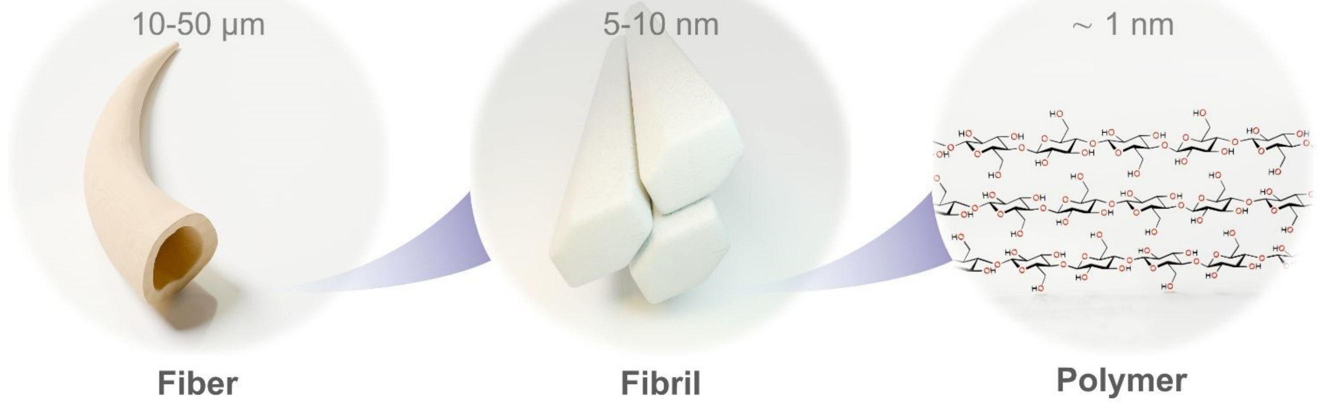

Figure 5: Multiple lengthscales in cellulosic materials, illustrated for wood-derived fibers: micrometric fiber widths (~10–50 µm), embedding nanoscale microfibrils (~ 5-10 nm wide), themselves composed of crystalline arrays of individual polymer chains.

Depending on the source and treatment, cellulosic materials can be obtained in various forms and at different scales, exhibiting hierarchical (nano)structural arrangements, from molecular chains to fibrils, fiber and bulk materials (Figure 5). The structure and properties being directly influenced by the level of organization.

At the macroscopic scale, one may work with large objects such as wood veneers or templates. [116, 131-133] At an intermediate scale, fibers constitute the most industrially relevant feedstock, available in established forms such as wood and plant fibers, kraft pulp or dissolving pulp all supported by mature pulp-mill infrastructure and competitive production cost (around 0.5-0.9 k$/t and 0.9-1.4 k$/t for kraft and dissolving pulp depending on grade and region). [118, 134, 135] Nonetheless, obtaining fibers with high-purity, high-DP cellulose is far from trivial, as conventional processing often involves harsh chemical treatments that can degrade cellulose chains and diminish performance. Reducing the size, often through more intensive processing, tends to yield more uniform and chemically pure materials. At the microscopic level, materials such as microcrystalline cellulose (MCC), like Avicel PH-101, widely used in academic research and pharmaceuticals, microfibrillated cellulose (MFC) or bacterial cellulose offer high purity and consistent properties while being already techno-economically viable in specialty application. [124, 136, 137] By contrast, nanoscale objects such as cellulose nanocrystals (CNCs) and the various forms of cellulose nanofibres (CNFs) are high-value materials with exceptional performance, but their current production routes remain comparatively energy- and chemical-intensive, which still constrains their deployment to higher-value, lower-volume applications. [61, 120, 138]

This diversity of sources and scales, while offering unmatched versatility, also brings significant complexity and non-uniformity (Figure 4 and 5). Harnessing cellulose effectively therefore requires strategic design tailored to the intended application. An additional layer of complexity arises when chemical modification is pursued, whether to tune intrinsic properties (e.g., hydrophobicity, processability) or to introduce new chemical functionalities.

Synthesis: Activation, Modification and Processing (II)

Tuning the properties of cellulose to broaden its range of applications often rely on chemical modification, for which several strategies are available (like targeting reducing ends or via oxidative cleavage). [139-141] But the most straightforward approach is to exploit the abundant hydroxyl groups, three per repeating AGU unit, present along the cellulose chain. [142, 143] Through these hydroxyls' derivatization, cellulose can be transformed into cellulose esters, ethers, sulfonates, and many other functional derivatives (Table 1). In principle, all the classical alcohol-based chemistry can be envisioned onto cellulose. However, a major challenge lies in the accessibility and reactivity of the hydroxyl groups, which are unevenly distributed within the supramolecular structure of the diverse cellulosic objects described above. Achieving high reaction conversions or high degrees of substitution (DS, defined as the average number of hydroxyl groups modified per AGU, ranging from 0 to 3) often requires strategies to activate or enhance the cellulose hydroxyls reactivity. [142]

The most direct way to increase accessibility is to dissolve cellulose completely.[144] Working under homogeneous conditions places cellulose within the familiar framework of post-polymerization modification. [145] All hydroxyl groups are equally available for reaction (though depending on their substitution level and position on the AGU). However, cellulose is insoluble in most conventional aqueous or organic solvents, necessitating the use of specialized and more exotic dissolution systems, which their precise dissolution mechanisms are still highly debated. [146, 147] Importantly, the solvents also need to be compatible with subsequent chemical modification. [90, 146, 148] Significant research efforts have been devoted to this challenge, leading to systems such as DMAc/LiCl, [149-152] alkali-urea aqueous media (e.g., NaOH/urea, LiOH/urea), [153, 154] imidazolium-based ionic liquids (BMIMCl, EMIMAcO, etc),[98, 155-157] DMSO/TBAF,[136, 158, 159] DMSO/DBU/CO2,[160-163], deep eutectic solvents (DESs), [164, 165] or superbase-derived ionic liquids.[166, 167] Within these systems, efficient derivatization, compatible with a broad range of reactions are possible, enabling higher and more uniform degrees of substitution usually under milder conditions than in classical heterogeneous protocols.[89, 90, 142, 168-172] At the same time, each solvent family comes with its own practical constraints such as sensitivity to water (DMSO/TBAF),[136, 158, 159] toxicity and recovery costs, and often narrow processing windows or high viscosity, which continue to motivate the search for more robust, scalable solvent platforms for cellulose chemistry.

Indeed, at the industrial scale, aside from fiber production (Viscose, Lyocell), cellulose derivatives are produced almost exclusively under heterogeneous conditions, primarily due to cost efficiency.[9, 173] To be more precise, they are often based on transient system, i.e. from heterogenous to homogenous, relying on prior activation or taking advantage of the later dissolution of the product. [174, 175] For example, cellulose ethers are typically produced by swelling and deprotonating cellulose under harsh alkaline conditions, followed by reaction with an alkyl halide (Table 1).[29] Or cellulose acetate, which is produced from high-grade pulp or fibers in a mix of acetic acid and acetic anhydride. [27, 174] The reaction, catalyzed by sulfuric acid, leads to soluble cellulose triacetate which is subsequently partially hydrolyzed to reach a DS around 2. This protocol ensures uniformity in the acetylation and better reaction control to target the desired DS. Furthermore, heterogenous conditions, even without leading to fully homogenous products, presents important advantages. [144] Even if the reaction is limited and/or confined to the surface, minor degrees of cellulose modification can lead to pronounced shifts in properties and enable access to new functionality.[176-178] Still, designing green chemical tools to facilitate chemical modification in heterogenous conditions remains an important target, and a task we are particularly interested in addressing. For example, we previously explored biobased ionic liquids, namely BBIL-AcO, as reactivity promoter in combination with DMSO or acetic acid for anhydride functionalization of cellulose. [179, 180] Moreover, heterogeneous processes are also often simpler to implement, facilitating product recovery without extensive purification, and allow retention of the material's native morphology.[178] This can be particularly advantageous when working with wood or fibers, where retention of the original architecture may help preserve mechanical performance.[181, 182] Finally, avoiding complex, fossil-derived, and potentially hazardous dissolution systems is also in line with the principles of sustainable and green chemistry.[144]

Sustainability and Green Chemistry (III)

An important aspect of sustainable materials development is the environmental footprint of the chemical processes used to create them. Using cellulose and other bio-based raw materials are logically seen as inherently attractive from a sustainability perspective, yet these benefits can be compromised if the associated synthetic pathways rely heavily on hazardous reagents, environmentally detrimental processes, or extensive use of fossil-derived chemicals. [144] Even though cellulose can be challenging to modify, its transformation should, as much as possible, adhere to the principles of green chemistry and sustainable development.[183-186] Developing systems that allow efficient functionalization, whether under homogeneous or heterogeneous conditions, while keeping the process simple and sustainable remains a significant challenge.[161, 162, 168, 187] Using the most relevant type of cellulose should also be taken into account as we discussed earlier: for example nanocellulose can be rather energy- and processing-intensive or associated with its own synthetic challenges.[188]

To remain relevant, we thus need to target simpler, benign, and ideally biobased solvents. [179, 189] In this context, water-based systems would be ideal or even solvent-free methods (e.g., ball milling). [190, 191] However, in aqueous media, the scope of reactions targeting cellulose hydroxyls is inherently limited due to the competing reactivity with water, despite notable examples involving oxime ligation,[192] silanization,[193] acetylation,[194, 195] and succinylation.[196] Water is also an issue in non-aqueous systems, as cellulose hygroscopic nature leads to strong retention of water molecules that are difficult to remove and leads to side-reactions and poor yields.[147, 156]

Thus, organic solvents are often needed, but the delicate balance between greenness, efficiency and compatibility with cellulose (or even cost) requires thoughtful and innovative design. Encouragingly, the past decade has seen substantial progress in this area. New ionic liquids systems through, for example, the development of superbase-derived ionic liquid or the elegant and switchable DMSO/DBU/CO_2 system has led to better recoverability, processing simplicity and overall efficiency. [160-162, 167, 197]

Structural Characterization and Analytics (IV)

As these processing strategies become more sophisticated, the demands placed on characterization and analytics increase accordingly. An essential challenge in cellulose research concerns therefore the comprehensive characterization and analysis of the material, an aspect much harder compared to common polymers. Achieving a comprehensive understanding of cellulose necessitates interdisciplinary expertise and consideration of phenomena across all relevant length scales, from the macroscopic down to the nanoscale (Figure 5). This spans from essential microscopy (optical, SEM, TEM, AFM), to scattering techniques (WAXS, SAXS, DLS, XRD), thermo-mechanical tests (tensile testing, DMA, DSC, TGA), and of course, chemical and spectroscopic analyses. One could argue that this multidisciplinary is common in polymer science. However, cellulose imposes an additional layer of complexity, particularly in chemical characterization, where heterogeneous structure and limited solubility significantly complicate analysis.

As discussed above, cellulose occurs in many solid forms and is far from trivial to dissolve. For that reason, most polymeric analysis requiring solutes, must be adapted specifically for cellulose. This is especially evident for two of the most critical tools in polymer chemistry: liquid-state NMR and GPC/SEC, where dissolution issues and non-standard protocols still pose a significant bottleneck to precise, high-value chemical characterization. Consequently, as cellulose science involves a broad range of disciplines, researchers often rely on simpler characterization techniques, which, while practical, may provide only partial information. FTIR, for example, is typically the first go-to technique because it is easy to perform, non-destructive, requires only small sample amounts, and is well suited to solids.[198, 199] It provides valuable information (e.g., presence of C=O, C=C, C–O, COO⁻, O–H, O-D groups) and is therefore a must-have for cellulose work.[200] However, FTIR alone is limited in providing in-depth chemical insight or confirming successful covalent grafting.

When more detailed analysis is required, other "non-dissolving" techniques such as conductometric titration or Raman spectroscopy can serve as valuable complements.[201-203] Titration enables quantification of ionizable groups (e.g., carboxylic acids), while Raman spectroscopy offers complementary vibrational information (e.g., for thiols) and can be combined with spatial mapping. [178, 181, 194, 204, 205] Additional solid-state techniques are particularly useful for cellulose, notably X-ray photoelectron spectroscopy (XPS) and solid-state NMR. [206, 207] XPS provides surface-sensitive information (sampling only a few nanometers) on elemental composition and chemical states, and is particularly powerful for monitoring surface oxidation, grafting reactions, and changes in the O/C ratio after functionalization of cellulose fibers or nanocelluloses. [178, 208, 209] Solid-state NMR, in turn, has been a critical tool, circumventing dissolution issues while providing insights into chemical composition, conformational arrangement, and structural organization.[210-214] Crystalline structure and chemical modification can be efficiently analyzed simultaneously with this technique. [195, 196, 215]

Nonetheless, solid-state NMR suffers from low resolution and sensitivity. This is precisely the advantages offered by liquid-state NMR. While fairly easy for some highly modified cellulose, made soluble in common deuterated solvents, performing such analysis for slightly modified or pristine cellulose is more complex. [26, 216] Recent advances have made this more accessible and reliable. A milestone development is the mixed DMSO-d_6/[P_{4444}][OAc] solvent system, which is potent, stable, and versatile for analyzing cellulose in many forms. In combination with diffusion editing, proper liquid-state NMR is made possible to perform advanced chemical characterization. [214, 217-220] Perfect complement to solid-state NMR, it can provide intel on fine chemical structures, covalent bonding, quantitative degree of substitution, regioselectivity, or possible side-reactions.[140, 194, 196, 217, 221]

The other major analytical pillar of polymer science, size exclusion chromatography (or gel permeation chromatography), has also seen constant improvements over the past decades. [151] While already a demanding technique for many polymers, it is particularly arduous for cellulose due to challenges associated with dissolution, aggregation or degradation during sample preparation. Nevertheless, it remains critical for determining molecular weight averages, distributions, and polydispersity, thereby complementing the chemical picture of a polymeric system. The DMAc/LiCl solvent system is most commonly used for this purpose, although alternatives such as DMSO-based systems are also possible. [151, 226-230] Still, expedient determination of molecular weight distribution remains a critical bottleneck.

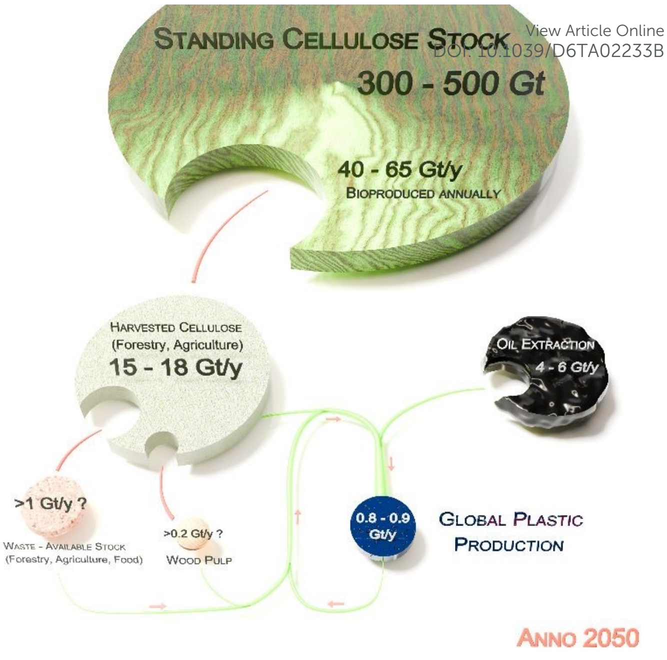

Figure 6: 2050 scales forecast and desirable circular shift of our material economy. Forecast for global plastic production[222, 223], crude oil [224 225] and potential HANPP (Human appropriation of net primary production) . [48]

Thus, taken together, these several challenges highlight a set of intertwined challenges: cellulose is difficult to process, its sources and length scales are highly heterogeneous, and its chemistry and structure remain harder to interrogate than those of conventional polymers. Yet recent advances in dissolution systems, heterogeneous and green modification strategies, and multi-scale analytical methods are progressively lifting these barriers and enabling more precise, high-value chemistries. As these tools mature, accelerating our understanding of cellulose structure and reactivity, they open the door to the next generation of cellulose-based functional materials.

Outlook of Cellulose for the next generation of Functional and Sustainable materials

The uneven historical trajectory of cellulose chemistry reflects not a lack of potential, but rather competition and comparison to petroleum-based polymers in terms of both cost and properties (Figure 1) As outlined in this perspective, direct substitution strategies has created a mismatch between the potential of cellulose, and the chemical strategies traditionally applied to it. Many of the challenges discussed throughout this viewpoint, including heterogeneity, insolubility, limited synthetic control, analytical difficulty, and different length and assembly scales, are often framed as fundamental limitations. From a sustainable materials perspective, these features should instead be regarded as defining characteristics that demand a

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different chemical mindset, one in which hierarchy and interfaces are treated as design elements rather than deviations from molecular uniformity.

This viewpoint requires a fundamental shift in how cellulose is conceptualised as a materials platform. Unlike synthetic polymers, cellulose is not a blank molecular canvas to be completely rewritten by chemistry; rather, its unique value lies in its pre-organised hierarchical structure, encoded through biosynthesis and assembly across multiple length scales. From this perspective, chemistry should not merely seek to overwrite this complexity, but to understand, preserve, and exploit it. With this in mind, we outline three complementary future directions that build upon and interconnect the challenges discussed above: two chemistry-centred viewpoints and one area that we believe warrants particular attention from the broader research community.

These include capitalising on hierarchical structure, developing polymerisation strategies compatible with cellulose environments, and establishing expedited analytical approaches and clear benchmarking frameworks. It should be emphasized that this is intended primarily as sources of inspiration and future research directions.

Harnessing Hierarchy for Function and Circularity (I + III)

Future progress will depend less on maximising degrees of substitution or enforcing homogeneity, and more on selective, spatially resolved, and function-driven chemistry that exploits hierarchical structure. In this context, interfacial and surface-confined reactions on fibres, fibrils, nanocrystals, or intact cell walls offer disproportionate leverage: they enable large changes in material performance and functionality through minimal, localized chemical intervention, while preserving, or even enhancing, the inherently superior mechanical properties embedded in cellulose assemblies (fibers, fibrils, wood). [194, 231] Recent developments in wood nanotechnology exemplify this approach, enabling high-value applications in sustainable functional materials.[116, 133, 232]

Leveraging the mechanical performance of cellulose-based materials effectively is a real compelling argument for adding value compared with common synthetic polymers. Examples of this emerging philosophy can already be found in several recently developed strategies. For instance, wood self-densification exploits the inherent hierarchical structure of wood to increase packing density and load-bearing capacity, achieving specific strengths of up to 422 $\text{MPa}\cdot\text{g}^{-1}\cdot\text{cm}^{3}$. [233-235] More generally, the use of high-DP cellulose and the preservation of high aspect ratios in cellulose (nano)fibrils can help maintain chain length thereby unlocking toughness (ca. 40 MJ m^{-3}) and strength (ca. 1 GPa) values widely unreachable for other common polymers. [135, 236, 237]

This shift in perspective also supports a broader reframing of circularity in cellulose-based systems. As illustrated in Figure 6, global plastic production is expected to double by 2050, while natural resources remain finite. Increasing cellulose extraction is not a viable solution, as it risks severe impacts on biodiversity and ecosystem stability.[48] Instead, circularity must extend beyond end-of-life depolymerisation or combustion towards a hierarchy-aware paradigm that includes reconfiguration, repair, reuse, valorisation, and chemical reconstruction of intact cellulose architectures. [238-240] Such strategies enable chemical recycling at higher macromolecular levels while preserving structural organisation and material value across use cycles, positioning cellulose as a genuinely circular materials platform rather than a disposable feedstock (Figure 6).[241] Within this framework, the balance between synthetic freedom and circularity must be carefully considered (Figure 7). The design of synthetic pathways and circularity strategies should be aligned with the targeted application and the intended material lifetime.

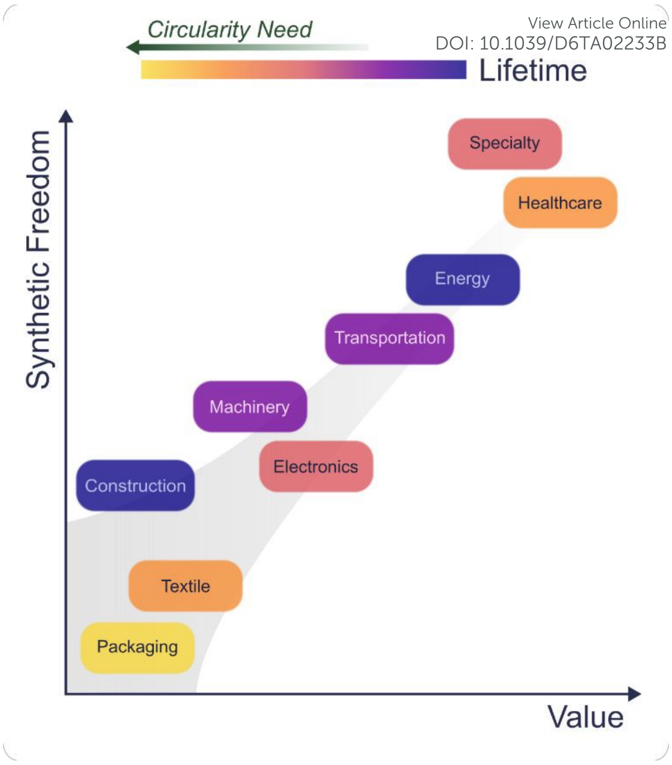

Figure 7: Considerations in chemical material design. Targeted application dictates synthetic freedom while expected lifetime drives the need for engineered circularity.

Historically, increasingly aggressive chemical modification has been used to access new functionality, often at the expense of reversibility, reprocessability, and long-term material value. Future cellulose chemistry must instead navigate a design space in which parameters such as degree of substitution, localisation of modification, and bond reversibility directly govern circularity outcomes. [204] Dynamic, reversible, and interfacial chemistries occupy a particularly favourable region of this space, enabling the introduction of functionality while retaining the capacity for reconfiguration and reconstruction. [242-244] To exemplify, recent advances in covalent adaptable chemistries provide new opportunities to impart reversibility, healability, and reprocessability, particularly in systems that must remain compatible with water and heterogeneous environments characteristic of cellulose-based materials.[245-247] Examples

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include water-confined chemical modification of cellulose, [194] as well as oxime and imine ligation strategies. [141, 192]

Polymerization Methods for Complex Environments (II)

It is important to keep in mind that the polymerization methods traditionally developed in polymer chemistry are largely rooted in petroleum-derived systems, and are therefore not inherently compatible with heterogeneous, water-rich, or fibre-containing environments. [248] Accordingly, an important frontier lies in developing polymerisation strategies deliberately tailored for compatibility with lignocellulosic feedstocks and the constraints imposed by intact hierarchical structures. [249, 250] Here, inspiration of biosynthetic strategies, e.g. the modular combinatorial radical polymerization of monolignols, is appealing due to its compatibility with water and oxygen. [251] Translating this to a synthetic concept could enable *in situ* growth of polymer networks or grafts without disrupting cellulose's hierarchical architecture, providing routes toward hybrid materials that combine native structural organisation with tailored chemical functionality. [252, 253] Here, the application space is immense: polymerisation pathways engineered to operate within the fibre environment open entirely new classes of functional cellulose-based materials, particularly in combination with degradable or chemically recyclable systems via modular polymerization strategies. [254-256] One particularly promising direction involves transparent biocomposites, where matching the refractive index of the matrix with the cellulose network enables optically clear, mechanically robust materials. [257-260] However, such approaches must be applied judiciously, as uncontrolled grafting or excessive polymer growth can disturb or obscure the underlying nanostructural features that give cellulose its unique mechanical and optical properties. [204, 253, 259, 261] Similarly, identifying and chemistries that remain selectively and predictably reactive in the presence of cellulose is crucial. Radical-based pathways are especially attractive: they can operate in heterogeneous, or fibre-rich environments, and work in water.[262] Recent work on radical grafting, fibre-compatible functionalisation, and heterogeneous polymer growth demonstrates how such chemistries can activate cellulose interfaces while preserving supramolecular integrity. [178]

Expedient Analytics and Clear Benchmarks (I + IV)

Even though analytical capabilities are constantly improving, they still represent a decisive bottleneck, as well as a major opportunity, in the ongoing transition toward more sophisticated cellulose chemistry. Many longstanding inconsistencies in cellulose research arise not from chemistry itself, but from insufficient, inappropriate, or misapplied characterization. As the field advances toward increasingly complex and partially modified systems, quantitative, structure-sensitive, and scale-aware analytical tools will be essential to correlate chemical modification with structure and performance across hierarchical cellulose objects. [263] In this context, establishing standardized protocols for evaluating cellulose chemistry—particularly for reporting degrees of substitution, regioselectivity, molecular weight, crystallinity, and surface chemical changes—would significantly enhance reproducibility, comparability, and scientific progress across laboratories and application domains. Taken together, these reassessments point toward a reframing of cellulose chemistry from a field historically focused on derivatization and substitution to one centered on deliberately adding interphasic functions into hierarchical biobased matter.

In summation, the long-term potential of cellulose lies not in competing with or replacing petrochemical polymers, but in enabling material concepts that are difficult or impossible to realize with purely synthetic polymers, including strong yet lightweight architectures, water-responsive systems, chemically tailored interfaces, and materials whose function emerges from structure across scales. In this sense, cellulose chemistry is not merely resurging; it is undergoing a qualitative transition. Its future relevance will depend on whether chemists can translate hierarchy, interphase modification, and dynamic bonding, alongside compatible polymerization methods into deliberate design strategies. By articulating both a long-term vision for cellulose-based materials shown in Figure 6 and the chemical design logic required to reconcile synthetic freedom with circularity shown in Figure 7, this Perspective aims to provide a framework that inspires the next generation of materials chemists to engage with cellulose not as a mere petrochemical substitute, but as an infinite canvas for materials creativity.

Author contributions

Lukas Marcos Celada: Conceptualization – Investigation – Visualization – Writing – original Draft - Writing – review & editing
Peter Olsén: Conceptualization - Funding Acquisition – Project Administration – Supervision - Writing – original Draft - Writing – review & editing

Conflicts of interest

There are no conflicts to declare.

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