Fulfilling the industrial potential of nanocellulose

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ABSTRACT

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Overpromising has been a constant trait of the nanosciences, and the case of nanocellulose makes no exception. Regardless of substantial academic and industrial research efforts re-started in the early 2000s, global production nanocellulose in 2018 amounted to less than 40,000 tonnes, chiefly in the form of microfibrillated cellulose for low value utilization in paper and cardboard products. Since the early 2000s market research analysts regularly estimate large market annual growth rates that so far never materialized. Besides replacement of conventional production methods with economically viable green chemistry processes, fulfilling the nanocellulose industrial potential requires to learn from the hype technology phase.

1. Introduction

Nanocellulose, namely cellulose fibrils with widths in the nanometer range [1], is a bionanomaterial endowed with truly exceptional chemical and physical properties that make it potentially useful in a wide array of industrial sectors [2]. Potential applications include advanced sectors well beyond paper making and cardboard packaging, to include enhanced membranes for fuel cells and water purification, aerogels, implant biomaterials, nanocomposites for car, boat and aircraft making, flexible electronics, stronger and more versatile coatings, and enhanced batteries [3].

In 2006 the research database Scopus indexed three studies dealing with nanocellulose (one book chapter, one conference paper, and one review) in the international scientific literature [4]. By the end of 2023, the same research database indexed 10,721 studies dealing with nanocellulose, of which 8,144 were journal original research articles, 1002 review articles, and 765 conference papers. Pointing to the global dimension of research in nanocellulose field, said studies were authored by scholars based in 119 different countries (the fifth of which, with 651 studies, is Malaysia) [4].

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Regardless of the aforementioned substantial research efforts, however, the market for nanocellulose remains small, with certain market analysts estimating it at $350 million in 2022 [5] and others at just $34 million [6]. Different market analysts, however, agree in predicting rapid market growth (compound annual growth rate of 20%), with the overall market estimated to reach $628 million by 2028 [5] and $1.1 billion by 2030 [6].

Yet, similar estimates published nearly on a yearly basis in the early 2010s largely failed to predict real outcomes. For example, in 2022 just 80 tonnes of cellulose nanofiber (CNF) were produced globally [7].

Analyzing the main technological and economic barriers to nanocellulose commercial uptake, Clauser and co-workers recently concluded that it is necessary "to develop new methods to produce new nanocellulose-based materials including environmentally friendly processes of extraction at a lower cost with reduced energy consumption processes" [8]. Put briefly, cellulose nanocrystals (CNCs) are extracted from cellulose by acid hydrolysis, microfibrillated cellulose (MFC) by mechanical treatment, and bacterial cellulose (BC) is microbially produced. Other critical challenges include the need to access low-cost raw material and to improve so far deficient coordination among government, industry, and academia [8].

As emphasized by the same team confusion in the marketplace also originates from a surprising confusion in nomenclature [8]. For example, MFC is often confused with CNF made partly oxidizing the cellulose primary alcohols via the 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO)-mediated de Nooy's process using bleach at alkaline pH followed by mechanical defibrillation. Using "cellulose nanofiber" as search query in a research database suggests that "studies on CNF began in 2004" [9]. On the other hand, Turbak and co-workers discovered MFC in 1977 and reported its discovery in 1983 [10].

In a key analysis on nanocellulose-related patents granted between years 1920 and 2011, Vázquez, Charreau and co-workers found that compared to MFC and BC, CNC

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(discovered in 1951 by Ranby and Ribi in Sweden using boiling aqueous H_2SO_4 2.5 N to extract the nanocrystals [11] consisting of highly crystalline cellulose I rod-shaped particles 50-400 nm x 5-10 nm that do not swell in water) gave place to the lowest number of patents, with their owners mainly being universities and public institutions [12]. However, growth in the number of patents was substantial for each nanocellulose form, with the years 2010 and 2011 having been the most prolific for each category. According to the same analysis, companies involved in nanocellulose field mostly originate from the pulp and paper industry.

In 2020, the team extended the analysis to nanocellulose patents granted between 2010 and 2017 finding an "astonishing increase" since 2010 [13], In detail, the scholars identified 4500 new patents, nearly 70% of which published in the last three years surveyed (2015-2017) [13]. Clearly, nanocellulose had become a hot topic of industrial research, with industrial interest unveiled also by the significantly larger share of companies being CNF patent assignees than amid those of CNC and BC. Again, most patenting companies were interested in CNF (~1700 patents) and BC (~1800 patents) rather than in CNC (~950 patents). Compared to the previous analysis of patents granted up to 2011 [12], along with Japan, China and the United States of America emerged as the most active countries in industrial research [13].

Overpromising has been a constant trait of the nanosciences [14], and the case of nanocellulose makes no exception. Besides replacement of conventional production methods based on the use of harsh chemicals or of energy intensive processes with economically viable green chemistry processes [15], fulfilling the nanocellulose industrial potential requires to learn from the hype technology phase. Lessons learned during said technology phase may indeed assist renewed managerial and applied research efforts aimed at large-scale utilization of this truly remarkable bionanomaterial.

2. Methodology

A search of overpromises and bold claims concerning nanocellulose was performed on the world wide web using different search engines. The web pages identified by the search engines included nanotechnology dissemination magazines, trade and scientific journals.

3. Results and discussion

3.1 Nanocellulose technology hype

Writing about nanocellulose identified as "a cheap, conductive, stronger-than-Kevlar wonder material made from wood pulp", Anthony in 2012 wrote on a widely read technology webzine:

> "In July, the US Forest Service opened the country's first nanocellulose plant in Madison, Wisconsin. This is only the third nanocellulose plant in the world, with the other two being in Canada and Sweden. The CelluForce factory in Montreal is now producing a tonne of nanocellulose per day. After a ramping-up period of a couple of years, the US Forest Service expects to sell nanocellulose for just a few dollars per kilo" [16].

The mentioned plant producing CNC in Canada with a capacity of 300 t/a was built in 2012 in Windsor, Quebec, on the site of a pulp and paper mill jointly owning the new company [17]. Ten years later, in 2022, the company reported annual revenue of $4,705,000 [18].

Nanocellulose, however, has never been "cheap". For example, even if a thorough cost manufacturing estimate in 2017 pointed to a manufacturing costs for CNC production "ranging from $3632 to $4420/t" [19], the price of CNC commercialized by the end of 2023 by a nanomaterial manufacturer based in Turkey varied between €5.4/g and €1.147/g, depending on the amount purchased (5 g or 1000 g, respectively [20]).

In a seminar aptly entitled "Cellulose nanofibrils (CNF) - a big hype or on the edge of a breakthrough" researchers based in Finland in 2015 concluded that CNF was "on the verge of a breakthrough" [21] and that "availability will soon" no longer limit applications even if some applications were "far, far away" [21]. Identifying "cost competitiveness" as the main challenge to nanocellulose practical applications, the same researchers highlighted the relevance of enzymatic fibrillation (using an enzyme cocktail comprised of cellulase enzymes with both endo- and exo-glucanase activity and minor hemicellulase activities) advanced at their Labs (HefCel, high-consistency enzymatic fibrillation of cellulose [22]) as a low energy input, low cost route to nanocellulose.

Research continued, and researchers at the same Finland's institution developed a compostable transparent film based on said nanocellulose combined with hydrophobic cellulose fatty acid ester "which looks and performs like plastic in food packaging" [23]. In mid 2022, the production of packaging material was in the pilot phase and its extensive industrial use was forecasted to take place "in 5-7 years" [24].

It is enough to conduct a search using the nanocellulose query at *nanowerk*, a website online since 2005 dedicated to nanotechnology, to identify 479 results [25]. In one spotlight article published on May 2023, Berger convened that:

> "The development of nanocellulose-based flexible materials is still in its nascent stages. There are significant challenges that need to be addressed before these materials can be widely adopted. Notably, nanocellulose is currently relatively expensive to produce,... current production methods are not equipped to meet the demands of large-scale production… additionally… nanocellulose is a highly sensitive material that can degrade easily… posing a substantial challenge to the development of durable, long-lasting nanocellulose-based materials" [26].

In another news, going back to 2020, an officer of Aalto University reported the news for which "Material manufacturing from particles takes a giant step forward" [27]:

> "The ability of nanocelluloses to bring together particles into cohesive materials is at the root of the study that links decades of research into nanoscience towards manufacturing... nanofibers extracted from plants are used as universal binders for particles to form a variety of functional or structural materials … As pointed out by the main author, Dr Bruno Mattos, 'This means that nanocelluloses induce high cohesion in particulate materials in a constant and controlled manner for all particles types. Because of such strong binding properties, such materials can now be built with predictable properties and therefore easily engineered'… [27]".

Another "spotlight" going back to 2013 concerning "Nanotechnology in the pulp and paper industry" [28] written by an officer of the Paper and Fibre Research Institute (PFI) readers were informed that:

> "One of the first scientific articles, which reported the advantages of a nanopaper concept for barrier applications,

was published by PFI and NTNU researchers. Due to the intrinsic strength of cellulose nanofibrils and their ability to self-assemble, dense and strong nanobarriers and nanocomposites can be manufactured…A necessary step to realize the above-mentioned applications requires the development of effective industrial production of nanocellulose, with affordable production costs. PFI and industrial project partners demonstrated that nanocellulose can be produced industrially, in large scale (1.5 tonne/day) and with low energy consumption (1600 kWh/tonne). This is considered a significant achievement and implies that nanocellulose from wood can be produced in large quantities, which is a major advantage compared to e.g. bacterial cellulose."

Recognizing "the potential and the hype" and noting that "forest companies" (*i.e.*, wood and paper mill companies) may have the right resources to produce nanocellulose in large amount, in 2014 Eklund published a Master thesis in business administration at Swedish University of Agricultural Sciences aimed at answering the questions: "why has the commercialization of nanocellulose not yet succeeded?" and "does nanocellulose hold a high market potential?" [29].

The advent of the internet, indeed, has led to a dramatic decline of paper (newsprint and printing and writing papers) demand [30], apparently originating the need for a wealthy and old industry such as the paper industry to develop new cellulose-based products such as nanocellulose.

Enquiring directly with Sweden-based wood and paper industries, Eklund found that nanocellulose production never started because nanocellulose was considered an additive to be used in existing or new products with little or no market demand ("demand is uncertain"). Wood and paper mill companies were found willing to support the forest bioeconomy. "Nevertheless", Eklund succinctly concluded "it has come to a point where everyone is willing to participate at the party but no one is willing to buy the ingredients needed for baking the cake - mainly because the guests' preferences are uncertain" [29].

In the subsequent decade, production of the bionanomaterial barely grew, but the hype surrounding nanocellulose continued unabated. In 2012 nanocellulose was termed "Nature's wonder stuff taking the world by storm" [31]. In 2018 it was identified as "one of the most promising green materials of modern times" [32], whereas in 2021 it had become "the next super versatile material for the military" [33].

3.2 Lessons learned

Bareis and co-workers have lately shown how, acting as attention-seekers riding "on the wave of attention" hypers in science and technology are "less interested in the long-term societal consequences of what happens when the wave collapses" [34]. Amid the long-term societal consequences of technology hype is investor disillusionment that, in its turn, will result in delayed (or permanent lack of) uptake of innovation.

It is therefore instructive to learn from successful innovators in the field of nanocellulose, namely from nanocellulose science and technology practitioners who succeeded in developing nanocellulose-based products demanded by the market. Though selling at \$50 g^{-1} [35] being made via inefficient fermentation methods using an expensive growth medium [36], bacterial nanocellulose for enhanced wound dressings so far is the most successful practical application of nanocellulose.

XCell, *Gengiflex*, *Membracel*, *Nanoskin*, *Securin*, and *EpiProtectt* are the tradenames of just a few BC-based wound dressings currently available on the market [37]. Serafica, a biochemical engineer, after his undergraduate studies in the Philippines moved to the USA where he got his doctorate in biochemical engineering and subsequently established his own company in 1996. Discussing the company's activities in various areas that contributed to the commercialization of the first two series of company's products [38], in 2016 he emphasized the unique nature of bacterial nanocellulose as a platform technology. The fermentation process indeed yields a very thin and pliable form, which is remarkably strong and yet gas and liquid permeable due to microporosity. Serafica underlined also the need for scientists to meet industry and innovate in order to solve real problems ("let the users decide the product for you, and become intimately associated with them because they will be the one to adopt it" [38]).

BC, for instance, was used by his company to treat venous leg ulcers with the added activity of accelerating the granulation and autolytic debridement of necrotic tissue with the fine, nonwoven, cellulose hydrophilic fibers comprising the multi-layered three-dimensional structure of bacterially synthesized *XCell* nanocellulose [39]; but also in neurosurgery as the first brain implantable product (brain patch) during a nine-year (2003-2012) development period that included prototype development, safety testing, and animal effectiveness testing costing about \$20 million [38].

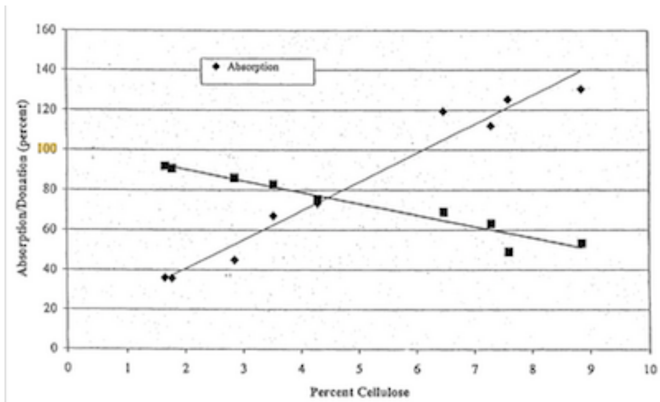

Figure 1. Absorption and donation capabilities of XCell microbial cellulose wound dressings versus the percent cellulose contained in the materials. All materials were of identical area and similar thickness. [Reproduced from Ref.40, with kind permission].

Having high strength and excellent biocompatibility the material is also able to either donate or absorb moisture (Figure 1, the region of intersection of the two curves shows the ideal cellulose content to maximize both properties [40]), depending on the wound environment, which makes it suitable for topical and implantable biomedical applications [41]. Eventually, in 2012, Serafica sold his company to a large pharmaceutical company that nowadays continues to successfully manufacture and commercialize the aforementioned BC-based healthcare products.

Besides a gift of the supercritical drying of microbial cellulose from a large paper company (and licensing from a large pharmaceutical company to Serafica's company the exclusive rights to its patents for using microbial cellulose in wound-care dressings) [42], these achievements were possible by learning how to shift from research to product development, while completing regulatory requirements and develop clinical experience to support subsequent product commercialization. Eventually, Serafica concluded, "the most difficult part of the process" [38] was in the selection of the right marketing partner as most new technologies "die" due to negligible market demand. Eventually, the third marketing partner was able to launch the product nationwide in the USA and, two years later, to commercialize it also in European countries so that after three more years a large pharmaceutical company purchased the whole business.

On the other hand, in early 2018 the Canadian forest nanoproducts network "ArboraNano" funded in early 2009, by the Government of Canada was dissolved after completing more than 25 research projects. After being dissolved ArboraNano gave to to FPInnovations (a not-for-profit R&D private organization which spans Canada's 180 pulp and paper industry, forest, and wood product companies) the task "to elaborate a fundamental research program to better understand the technological gaps" [43].

In brief, whereas the bottom-up approach based on nanocellulose bacterial synthesis has been widely commercialized, two decades of intense applied researches so far were not enough to solve the main problem that so far delayed massive nanocellulose manufacturing by the top-down production routes.

For example, Nippon Paper, which operates a 500 t/a CNF plant in Ishinomaki based on the TEMPO oxidation process, in mid 2023 announced its intention "to soon put the new process into practical use in a bid to lower production costs referring to process capable to convey CNFs dispersed in water" [44]. According to a company's research manager "samples of CNFs once sold for as much as 100,000 yen (around $750) per kilogram" [44]. The firm's objective in mid 2023 thus was "to get prices down to tens of thousands of yen" because "if prices can be squeezed to a few thousand yen, the market will grow dramatically" [44].

Japan, where a "Nanocellulose Forum" was established in 2014 to strengthen cooperation between industry, academic research, and government consolidated in 2020 into "Nanocellulose Japan", is the world's leading country in terms of industrial utilization of nanocellulose [45]. In 2017, sports shoes whose midsoles are strengthened and their weight reduced by adding CNF (cellulose nanofibers are light and elastic but, like glass, show low thermal expansion and contraction) to the polymer, were the first commercial product containing CNF. In six years, the sports shoe company sold 10 million pairs of said high-performance shoes [44]. Alas, the high performance auto tires whose rubber is functionalized with CNF for enhanced durability and ride comfort commercialized in 2019, today are no longer sold.

Japan-based researchers have already developed not-degrading gears made of polyacetal reinforced with CNF that are never worn away or wear away counterparts sliding on each other without lubricants; concept car that weighs about 15% less and consumes 11% less fuel than a conventional car of comparable size (presented in 2019); as well as reinforced plastic in which glass fiber is replaced by CNF affording CNF-reinforced plastics of exceptional degree of recyclability (when CNF-RP is recycled, the cellulose nanofibers do not break so that the mechanical and thermal properties are not degraded every time the plastic is recycled) [44].

From reinforced paper and board through sporting goods, vulcanized fiber, building materials, paints, audio equipment, personal care, cosmetic and food products, the list of products using CNF commercialized in Japan [46] shows evidence of the often-claimed "exceptional versatility" of nanocellulose.

We have recently suggested that, along with low production cost, the two main drivers for successful bioproductions are the high price of the alternative, oil-based target product and access to readily available low cost aw material [47]. Production of CNC via sulfuric acid hydrolysis is both capital intensive and has high operating costs even in the lowest cost plant configuration (without acid recovery), with feedstock cost and capital investment being the major cost drivers [19]. On the other hand, the price of transparent ethyl vinyl alcohol (EVOH) copolymers currently widely used in packaging for food preservation following the outbreak of COVID-19 climbed from \$5.5/kg in 2019 to \$11-\$14/kg at the end of 2022 [48].

This creates room in the marketplace for CNC-based transparent coatings, well-known for their excellent oxygen, and oil and grease barrier properties, especially when extraction of CNC (using sulfuric acid at high concentration) takes place from the sludge produced by pulp and paper mills and not from expensive microcrystalline cellulose or bleached pulp. Indeed, a company based in Israel jointly owned by a large paper company based in Sweden established in 2010-12 originally targeting the production of "ecologically friendly foams", recently commercialized a product available as water-based coating, that allows for the recyclability of plastic food packaging [49]. Actually, the coating can be applied also to cosmetic, pharmaceutical and agriculture packaging plastic products using existing standard industrial coating machines.

The latter feature of the technology is also important because industry is not willing to face substantial new investment in new coating devices to make their packaging sustainable, namely recyclable or reusable moving away from single-use plastics. The company, which operates a pilot-scale production line producing up to 100 kg of sulfated CNC per day, was recently forecasted to be able soon "to manufacture 200 tons of CNC per year in Sweden" [50].

4. Conclusions

The study of technology hype concerning nanocellulose has plentiful lessons to teach. Said lessons are not only useful to bioeconomy company managers but also to researchers as "the excitement that currently surrounds nanocellulose research" [51] may actually vanish and be replaced by skepticism due to misleading and subsequently unmet expectations as it happened for many nanotechnologies [52].

Forecasts dating back to 2014 for which the market of nanocellulose has a potential of 1.75 million tonnes globally only for low volume applications (aerogels, wallboard facing, special paints, aircraft structure and interiors) and of 33 million tonnes for high volume applications (packaging coatings and fillers, paper coatings and fillers, automotive structure and interior, clothing textile and cement) [53] should be compared to reality. In 2018, the nanocellulose market amounted to 39,600 t, with the largest share being made of MFC produced at pulp mills to functionalized paper and cardboard products [54].

The aforementioned estimates were based on the assumption made in 2013 that cellulose nanomaterials can reach the price target of \$4.4/kg to \$11/kg [53]. As noted above, the research manager based at Nippon Paper (*i.e.*, the company operating the world's largest CNF manufacturing plant) in mid 2023 was still referring to his company striving for CNF "once sold for as much as 100,000 yen (around $750) per kilogram" [44] aiming to "to get prices down to tens of thousands of yen" [44].

Remarkably, in sectors where the high $50/g price of (bacterial) nanocellulose is not an issue such as in the case of wound dressing or in neurosurgery as the first brain implantable product (brain patch), the market uptake of nanocellulose took only a few years (less than a decade, in the case of *XCell* [38]). On the other hand, in sectors where nanocellulose competes with widely employed and lower cost oil-derived products, nearly no commercial uptake of nanocellulose took place in over two decades of commercialization efforts.

So-called "forest" (wood and paper pulp) companies, namely so far the main investors in nanocellulose research and production plants [12,13], should be aware that their main competitors are chemical industry companies that produce nearly every polymer and fiber material from oil-derived feedstocks using highly efficient, continuous catalytic processes. This implies, as detailed elsewhere [47], that successful productions of nanocellulose fibers will be those targeting the production of expensive nanocellulose-based products in relatively high demand (for example the strong, expensive carbon fibers used in aerospace and luxury vehicles) preferably from low cost cellulosic raw material (*i.e.*, not microcrystalline cellulose, bleached pulp or cotton).

Eventually, the large-scale uptake of cellulose nanofibers in widely different industries in place of synthetic organic polymers for the high volume end uses only will happen when the existing nanocellulose production processes will be replaced by greener and significantly less energy-intensive processes allowing the long-awaited manufacturing cost reduction. Amid said processes, Karimi's and Pagliaro's teams have lately identified mechanochemistry ball milling, the one-pot heterogeneously catalyzed oxidation process over solid TEMPO-based catalyst, enzymatic catalysis, and cavitation (both acoustic and hydrodynamic) as the four most promising green chemistry routes to cellulose nanofiber or cellulose nanocrystals [15].

In conclusion, willing to answer the question "who should be interested and why?", researchers and companies developing new products from nanocellulose should be aware that a number of new processes to produce bacterial nanocellulose, CNC and CNF with high yield and at substantially lower cost have been developed, that allow for vertical integration (namely an arrangement in which the supply chain of a company is integrated by the same company) [15]. Others will most likely emerge.

Work on nanocellulose and research is indeed progressing rapidly. Pointing to unprecedented global interest, in the first fifteen days of 2024, a comprehensive research database already indexed 215 papers in peer reviewed journals (or selected preprint platforms) published in 2024, of which 194 were research papers [55]. Out of said 215 papers, 104 were authored by researchers based in China. This is relevant, because, from silicon solar cells through Li-ion batteries and light emitting diodes, China's industrial system is well-known for its ability to rapidly turn fundamental knowledge into successful industrial productions.

Putting the hype surrounding nanocellulose in the first two decades of the 2000s in the broader context of exaggerated promissory claims typical of nanotechnology [14,52,56], this study investigates technology hype concerning nanocellulose in the context of overpromising in the nanosciences. Its outcomes will hopefully accelerate progress towards large-scale production and commercialization of this versatile biomaterial.

Competing interest

The Authors declare no conflict of interest.

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