Perspective

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Tannin: a new insight into a key product for the bioeconomy in forest regions

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Abstract: Produced by extraction from certain woods and barks with boiling water, tannin is the name given to a mixture of high molecular weight biophenols increasingly used in a number of industries. This study offers a new bioeconomy insight into an old natural product that, we argue in this study, will likely play a crucial role in the development of the bioeconomy of forest regions. After providing an updated picture of selected key economic and production aspects, we show how flourishing research on tannin's biological activity and technological applications has revealed many new properties that are likely to drive significant growth in demand in the near and mid-term future. The study concludes with two lessons learned and one forecast that will hopefully accelerate progress in the bioeconomy of forest areas based on the circular economy of tannin enabled by new extraction and purification technologies. © 2021 Society of Industrial Chemistry and John Wiley & Sons Ltd.

Key words: tannin; forest regions; bioeconomy; polyphenols; chestnut; circular economy

Introduction

Deriving its name from the Latin word *tannum* for 'oak bark', tannin is the name given to a mixture of oligomeric polyphenolic compounds widely employed in the leather, wine, food and beverage, chemical, cosmetic, pharmaceutical, and mining industries.[1] The biophenols of tannin combine with collagen and other proteins contained in animal skin transforming it into chemically and biologically stable leather due to the inhibition of proteolytic enzymes.[2]

Excellent recent books[1] and reviews[3] have been devoted to tannins, including an account of their biological (antioxidant, anti-inflammatory, antidiabetic, cardioprotective, wound-healing, and antimicrobial) properties.[4] Pizzi, a pioneer in research on tannin-based adhesives, has recently explained how three subsequent phases characterized the economy of the tannin industry.[5] The first started around 1850 in Lyon, France, with the production of iron tannate for the black coloring of silk for women's blouses, followed, a decade later, by the uptake of tannin in place of oak chips from the leather

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industry. Finally, after World War Two tannins were first adopted in the production of bio-based adhesives, and, more recently, in composite materials.

Driven by the use of tannins in the preparation of adhesives and resins, in the early 1980s, the tannin industry had already shifted from hydrolyzable tannins (mixtures of simple phenols, such as pyrogallol and ellagic acid, with gallic and digallic acid glycosides) to condensed tannins (condensed flavans containing no sugar residues). In 1982, the latter already constituted over 90% of the global production (~350 000 t) of commercial tannins.[6]

This study offers a new insight on tannin as a key product of the emerging bioeconomy in forest regions. The bioeconomy is the economy in which materials and functional substances are obtained from biological resources, while useful energy (electricity and heat) is obtained from renewable energy sources, particularly sunlight, wind, and water.[7]

First, we provide an updated outlook on key economic and market aspects often absent in scholarly papers. Then, we focus on innovations in tannin applications driven by flourishing research that might soon lead to a further large increase in demand.

The study concludes with two lessons learned and one forecast that will hopefully accelerate progress in the bioeconomy of forest areas based on the circular economy of tannin enabled by new extraction and purification technologies.

Economic and production aspects

Produced in Europe, the Americas, Africa and Asia generally by extraction of selected woods and bark (including oak, birch, quebracho, mimosa and chestnut trees) with hot water, tannin is generally supplied to customers either as a brown powder or as a brown-black 50% concentrated aqueous solution.

According to a reputed market intelligence company, in 2017 the global tannin market size was estimated at 1.1 million tonnes, with four main applications: leather tanning (accounting for over 62% of the total market revenue in 2016), followed by wood adhesives, wine production, and anticorrosive primers.[8] The market, expanding at an estimated compound annual growth rate (CAGR) of 6.7% was forecast to reach $3.39 billion by 2025.

According to another market intelligence company, publishing the most recent (February 2021) market report, the tannin market size in 2020 was assessed to be 1.4 million tonnes, growing at an estimated CAGR of 5.5% over the period 2020–2027.[9] An anonymous reviewer for this manuscript commenting on these figures, added:

I am afraid that the total production of commercial tannins in the world today, both hydrolysable and condensed tannins, is not more than 220–230 thousand tonnes per year.

This has been the limiting factor for their utilization, notwithstanding that there is a lot of interest in them at present. The amounts you quoted from reputed market analysts do not refer to the actual production… but refer to the potential production if more factories to extract tannins are built. Yes the potential is enormous, even more than what you quote, but it is not realized.[10]

This discrepancy may be due to figures referring either to pure (100%) tannin, or to tannin extracts in which the actual amount of tannin in the powdered or liquid extract is a fraction amid several components.

Indeed, the industrial extraction process relies on the leaching of water-soluble tannin from wood or bark in hot water. In brief, chips of wood trunk or bark are loaded in large autoclaves. Typical botanicals sources are black mimosa bark (*Acacia mearnsii*), quebracho wood (*Schinopsis batansae*), oak bark (*Quercus* spp.), sweet chestnut (*Castanea sativa*), and pine (*Pinus radiata* and *Pinus nigra*) woods.

The extraction uses hot water and lasts about for about 6 h,[11] Generally, a 4–5 wt% tannin solution is obtained, with an extraction yield of about 60–65%.[12] After clarification by settling, the tannin solution is concentrated in multiple evaporators by evaporation under vacuum to limit the tannin oxidation, up to the desired concentration (>50 wt%).

For example, one company in Brazil was reported in 2017 to use water at 75–85 °C in a series of autoclaves performing a countercurrent process to extract tannin from the bark of mimosa. The extracted liquor is concentrated and air dried to afford a finely powdered tannin extract chiefly consisting of condensed tannins (polyflavonoids, 70–80%). The residual components of the powder are hydrocolloid gums, sugars, and other small-molecule substances.[13]

Used since the industry's early days, the tannin extraction process saw only one significant change until the late 1990s, when the boiling pools were replaced with counter-current extractors. Chopped wood or bark is loaded into a battery of autoclaves working in counter-current.

Tradenamed 'Tantec', an improved extraction process requiring significantly lower energy and of higher versatility was commercialized in the late 1990s. Extraction takes place again via leaching of wood or bark tannin in hot water. In place of evaporation, however, concentration now takes place via nanofiltration through polymeric membranes (Scheme 1) affording a 50 wt% tannin aqueous solution.[14] If needed, a dried powder is obtained via spray drying.

In contrast with the conventional process, steam or hot water is used only in the extraction step. During the

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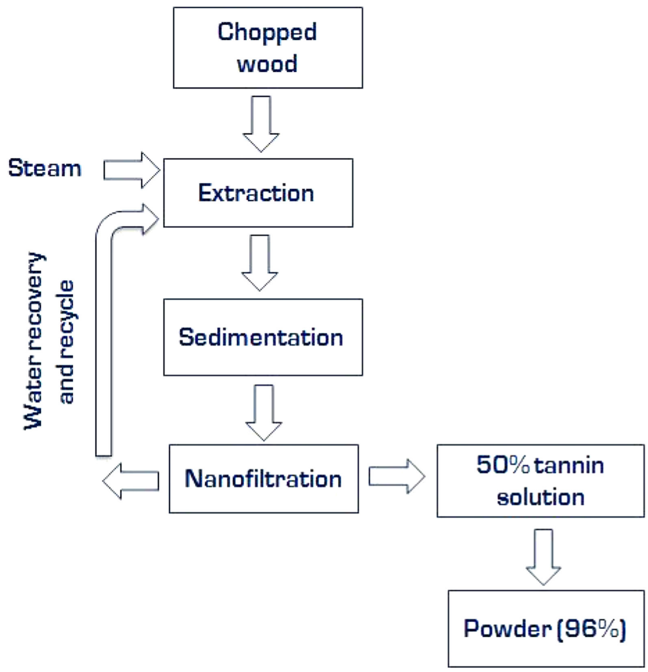

Scheme 1. The Tantec tannin extraction process (adapted from ref. 14, with kind permission).

crucially important concentration step, the temperature does not exceed 40 °C, with a significant improvement to the occupational health and safety conditions.

The same nanofiltration can be used to recover tannin from exhausted tanning liquors from the leather industry, affording a 75% reduction in chemical oxygen demand of the effluent, with a direct decrease in wastewater disposal costs, saving water and energy.[15]

In general, tannin extraction plants across the world continue to use the solid–liquid extraction process, employing large amounts of hot water followed by evaporation under reduced pressure, although numerous alternative processes, from microwave- to ultrasound-assisted extraction, on a laboratory scale, have been shown to afford higher tannin yield and reduced extraction times.[16]

No acid, alkali or enzyme is used in the extraction process. However, the process is highly energy intensive due to the large specific heat capacity of water and the 2.4 water-to-solid weight ratio typically employed. Chipping, furthermore, typically consumes 15 kWh/t of electricity.

Emerging applications

Flourishing research on tannin's biological activities and technological applications has revealed many new properties, which have expanded the demand for tannin even to the mining industry, where tannin is used to separate and concentrate metal compounds selectively in ores and other complex mixtures of materials. Applications range from coagulants in the potabilization of surface water and in the treatment of industrial wastewater through to dying agent for natural textile fibers.[17]

To understand the environmental benefits of using biobased tannins in place of heavy metals or synthetic molecules, it is enough to consider that leather tanning using vegetable tannin replaces tanning with toxic Cr(III) salt, which results in occupational exposure to Cr(III) in organic solvents or in protein bound form (leather dust).

The latter treatment causes serious health and environmental hazards in many areas of the world hosting leather companies.[18] On the other hand, treatment of raw hide with vegetable tannins combined with aluminum sulfate is able to produce leather nearly identical to that obtained using chromium-based tannin.[19]

In the following we focus on two new applications of tannin that might lead to a large growth in demand in the near future and in the mid-term (5 to 10 years).

Chestnut tannin as biopesticide and plant corroborant

The first new application, already commercialized following studies started in Italy in the early 2000s, concerns the use of vegetable tannin as plant corroborant – namely enhancing plant resistance to harmful organisms, and improving radical development by reducing damage due to parasites.[20]

Tradenamed Tannisol PB, a standard chestnut tannin powdered extract with a 75% tannin titer obtained via an extraction-nanofiltration process is commercialized as a registered plant corroborant at > €9/kg when bought in 25 kg batches (otherwise >€12/kg for a 1 kg batch).[21] For comparison, the price of tannin reported by a research team based in France (a leading tannin production country) in 2015 varied between €0.7/kg and €1.5/kg depending on the purity and on the botanical resource.[3]

Approved in organic agriculture, the product is distributed by 'fertigation' during the crop development cycle in the last phase of the irrigation cycle (to avoid leaks due to leaching) on fruit and vegetable crops. The product, however, is also a biopesticide (although not registered as such) because it exerts a broad scope antimicrobial and antifungal action by inhibiting the hydrolytic enzymes (cellulases, pectinases, xylanases) used by many pathogens to penetrate plant tissues. This effect adds to its repellent action against predator insects and parasites, due to altered plant tissue flavor (astringency) and hardness.

The latter properties were investigated in plants in a EU-funded project between 2014 and 2016, showing evidence that the chestnut tannin extract promotes both a more vigorous plant growth, while acting as pesticide against phytopathogenic bacteria on crops such as kiwi and nematodes typical of tomato and tobacco plants.[22] Remarkably, ecotoxicity assays showed that all polyphenol extracts studied, including tannin from sweet chestnut, were free of toxic effects even at the concentration of 1 g/L, thus enhancing the agronomic and environmental relevance of this new biopesticide and plant corroborant.[23]

Tannic acid for Li-ion battery cathodes

The second application, not yet commercialized, concerns the use of tannic acid, a low molecular weight polymer of gallic acid and 3-galloylgallic acid esterified with glucose obtained by extraction of the wood of *Castanea sativa* (sweet chestnut) and *Quercus infectoria* (Aleppo oak), for producing the most important component of Li-ion batteries, namely the cathode.

In early 2021, researchers in Germany reported that it was enough to ball mill tannic acid and porous carbon, easily derived from sucrose, to form a stable composite (C/TAN) showing excellent capacity of 108 mAh g^{-1} at 0.1 A g^{-1} and low-capacity fading.[24] Tannic acid, via the quinone-catecholate redox mechanism, and porous carbon, through its high surface area and excellent conductivity, are responsible for the high capacity observed.

n detail, the capacity using charging-discharging tests during long-term cycling raises during the first 17 cycles, and slowly decreases afterward (Fig. 1), with a capacity retention of 81.1% after 90 cycles. With approximately 3.4 V versus Li^+/Li, this cell features one of the highest reversible redox potential reported so far for biomolecular cathodes.

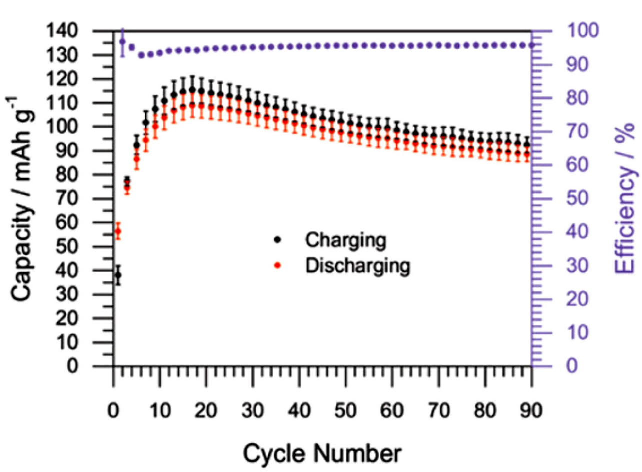

Figure 1. Electrochemical performance of C/TAN: charging–discharging test at 0.1 A g^{-1} (reproduced from ref. 24, with kind permission).

Ball milling ensures good contact between tannic acid and microporous carbon, facilitating the formation of π–π stacking between the microporous carbon and tannic acid, preventing dissolution of the otherwise highly soluble tannic acid (Fig. 2(b)), whereas the porous structure of the cathode benefits the intimate contact between the heterogenized redox-active tannic acid and the liquid electrolyte (1.0 m $LiPF_6$ dissolved in 50:50 ethylene carbonate/diethyl carbonate), leading to fast reaction kinetics.

In other words, tannic used in a composite with porous bioderived carbon to form the cathode of a Li-ion battery achieves a high voltage, solving at the same time the main drawbacks of organic electrode materials based on the carbonyl (C=O) reaction based on the reversible electrochemical reaction between lithium ions and carbonyl groups in a conjugated organic framework (Fig. 2(a)), namely the low potential as cathodes, the low electronic conductivity, and the high solubility of the redox-active organic material in the electrolyte.[25]

The cathode, composed of lithium ion 'intercalating' crystalline inorganic oxides, such as lithium ferrophosphate or lithium nickel manganese cobalt oxide, is the most expensive and important component of the Li-ion battery (LiB).[26]

Replacing the inorganic oxides mentioned above used to manufacture today's LiB cathodes with a readily available organic material such as the tannic acid/carbon composite also mentioned above would not only dramatically lower LiB production costs (an ongoing trend due to massive deployment of LiB production factories in China),[26] but would also lower the market entry costs because the material is readily prepared by the safe and simple milling of two powders.[24]

Outlook and perspectives

Two main lessons and one forecast of relevance for bioeconomy researchers and for policy makers based in forest areas originate from the present insight into the bioeconomy of tannin, an old forest product manufactured on an industrial scale for almost two centuries.

First, tannin has an exceptionally versatile chemistry and biochemistry, which will drive applications in numerous other fields, well beyond today's four dominant uses (leather tanning, wood adhesives, wine production, and anticorrosive primers).

Second, likewise to what happened with the rediscovery of the precious role of phlorotannins in protecting marine oils from oxidation and autooxidation,[27] bioeconomy scholars find in the case study of tannin a clear example of

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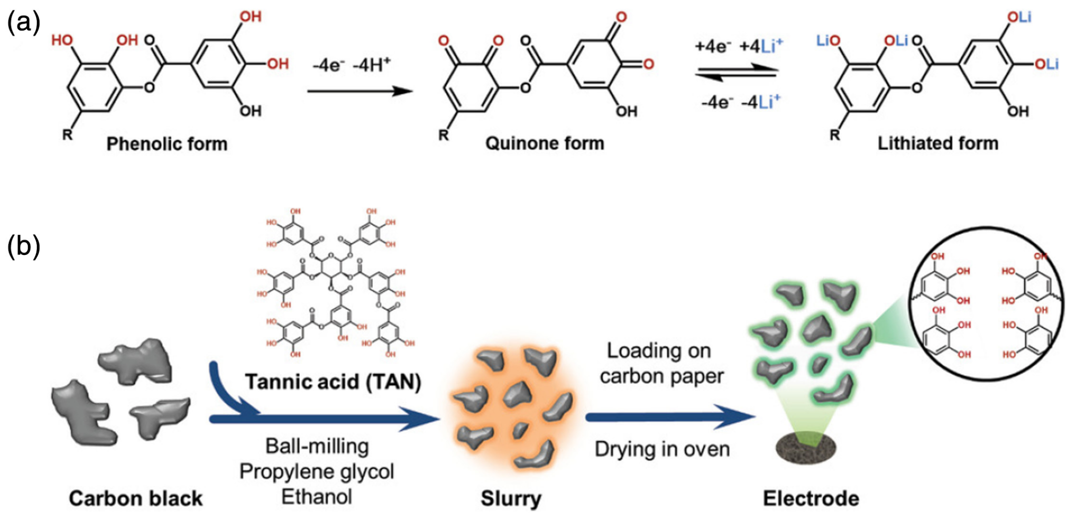

Figure 2. (a) Tannic acid has a high density of redox-active o-hydroquinones. (b) Ball milling mechanism affording the C/TAN composite (reproduced from ref. 24, with kind permission).

how progress in science and technology is far from being linear.[28]

Driven by advanced usages, we anticipate that new green chemistry production and purification technologies[16] to produce high quality tannin extracts will be likely industrialized in the near future.

One such new application of tannin is as biopesticide and plant corroborant, a field pioneered in Italy.[20] Another could be in Li-ion batteries, as recently demonstrated at laboratory scale in Germany.[24]

If and when production of Li-ion battery cathodes based on tannic acid commences, which would require them to demonstrate prolonged stability in storage, delivering electricity at the original high voltage (a typical LiB using a $LiFePO_4$ cathode lasts 10 years or more if charged and managed correctly), a supply of high purity tannic acid currently sold by chemical suppliers at \$200–300/kg[29] will be required.

Tannin production plants located in multiple countries, including France, Brazil, India, Japan, the USA, Zimbabwe, Tanzania, Kenya, Argentina, Slovenia, Italy, Canada and Switzerland, currently mostly use the old production route based on extraction with boiling water using counter-current extractors.

Aware of the new as well as of the emerging uses of tannin in advanced technologies, today's and tomorrow's tannin manufacturers will progressively adopt new extraction and concentration technologies, which have so far been confined to the laboratory scale.[16] Eventually, local communities and indigenous peoples of forest zones will benefit from increasing revenues derived from increased production of tannin, and from reduced environmental impact of tomorrow's tannin production plants which will rely on the aforementioned green extraction and green purification technologies.

Likewise to what happens with overfishing and the need to increase the revenues of fisheries through valorization of fish leftovers via new green extraction technology,[30] the latter is a crucial factor to arrest and prevent deforestation. New revenues and new jobs related to the production of tannin would provide new financial resources supplementing the income of residents in forest areas, thereby establishing a realistic alternative to cutting more trees for generating new revenues.

This is especially relevant considering that tannin can be successfully extracted from the bark residue of the timber industry, as in the case of the bark of *Pinus pinaster* (maritime pine), rich in procyanidin condensed tannins.[31]

Rather than relying 'on false promises made in the interest of profits of powerful transnational companies as found in an interesting analysis of alleged "value-free" discourse of EU and US governments on the bioeconomy',[32] the social and economic benefits of the bioeconomy of tannin are already evident, with a diffuse global tannin industry and market due to the presence of numerous regional and domestic players in different countries.[5,8,9]

Tannin, a forest bioproduct sold in 2015 at \$1.5/kg,[3] in advanced applications such as the uses mentioned above as an environmentally friendly agrochemical, is sold at prices approaching \$10/kg for large orders, and well beyond \$12/kg for small scale use, as in protecting garden plants.

Beyond numerous new jobs for plant workers, significant professional opportunities will originate from the evolving tannin industry: in research (for life scientists, chemists, and agronomists), in designing and running the new tannin production plants (for engineers and technologists), and in managing the existing and the new tannin production companies (for managers and for marketing managers).

This study will hopefully contribute to accelerating this progress.

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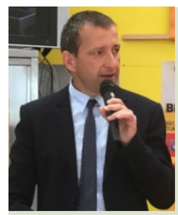

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