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Review

Nanocellulose Membranes for Fuel Cells and Electrolyzers: A European Perspective

Published as part of Energy & Fuels special issue "Novel Routes to Green Hydrogen Production in Europe".

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Cite This: *Energy Fuels* 2026, 40, 4445−4451

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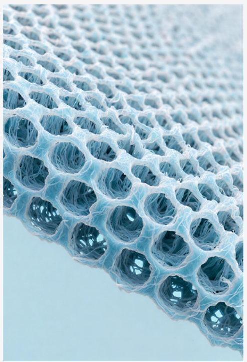
ABSTRACT: Researchers based in different European countries are actively developing nanocellulose membranes as sustainable alternatives to conventional membranes used in both hydrogen fuel cells and anion- and proton-exchange membrane electrolyzers. Focusing on recent innovations, this study offers a European perspective on research on nanocellulose-based ion-exchange membranes for new-generation hydrogen fuel cells and water electrolyzers. The study highlights also how research policy promoting a cross-disciplinary and cooperative approach to research enabled the innovation behind the development of these enhanced membranes. Perspective is timely as research efforts conducted in European and in non-European countries in the past decade (2015−2025) are approaching technology commercialization.



1. INTRODUCTION

"Green" (or solar) hydrogen obtained from water via electrolysis powered by electricity obtained from renewable energy sources, particularly wind and sunlight, is a critically important clean energy storage technology.[1] Hydrogen thereby produced can be recombined with air's oxygen in fuel cells generating electricity and heat. Due to quick refilling and high-energy density of H_2 compressed at 300 bar, green hydrogen stored in today's safe cylinders in composite material is particularly well suited to power heavy-duty fuel cell electric vehicles such as trucks, buses, trains, ships, and even airplanes.[2]

For widespread uptake of the technology, the production cost of "green" hydrogen via the electrolytic process, requiring a relatively large amount of electricity (55 kWh/kg H_2), needs to be reduced from today's \$5−6/kg to less than \$2/kg. For example, in the USA, the Department of Energy in 2024 aims for green hydrogen production costs of \$2/kg by 2026 and \$1/kg by 2031.[3]

Today's green hydrogen is chiefly sourced via alkaline water electrolysis (AWE) mediated by low cost and durable nickel-based electrodes in electrolysis cells operated at 80 °C in 30% KOH.[4] This old, yet important technology is employed to produce less than 5% of global hydrogen output. In 2021, for instance, around 4% of the global hydrogen production was obtained via electrolysis of water (almost 47% from natural gas, 27% from coal, and 22% from oil as a byproduct).[5]

Though more expensive and working under acidic conditions, proton-exchange (or polymer electrolyte) membrane (PEM) water electrolysis, in which a proton-exchange polymeric membrane acts both as a gas separator and an electrolyte for the selective proton transport, is the most efficient water electrolysis technology.[6] Integrated between the anode and cathode, the ion-selective membrane in fuel cells and electrolyzers is the key component that allows transport of specific ions (typically protons or hydroxide) blocking molecules such as O_2 and H_2 consumed or generated at the electrodes.

Reduction in cost of electrolytic hydrogen requires also the development of new advanced membranes of lower cost and enhanced performance for both acid and alkaline water electrolysis.[7] Highly conductive, chemically stable, and mechanically robust electrolysis membranes are urgently required. In the case of PEM electrolyzers (and PEM fuel cells), this requires improvement of membrane durability.[8]

Today's state-of-the art proton-exchange membranes having the highest performance in terms of conductivity are made of perfluorosulfonic acid (PFSA) polymers.[9] Mechanical and chemical degradation, however, progressively reduce the performance of PFSA membranes over multiple working cycles due to local changes in relative humidity and mechanical stress

Received: December 15, 2025
Revised: February 5, 2026
Accepted: February 6, 2026
Published: February 16, 2026

https://doi.org/10.1021/acs.energyfuels.5c06561

leading to reduced interfacial contact between the membrane, hole formation, and membrane thinning.[10] Moreover, proton conductivity at temperature exceeding 80 °C in PFSA membranes is no longer observed at relative humidity (RH) levels <50%, requiring RH values >60% to drive proton transport thanks to higher amount of more H-bonded water networks.[11]

Chemical degradation, in its turn, is driven by polymer chemical decomposition as $^{\bullet}$OH radicals generated during PEM electrolyzer or fuel-cell operation attack the polymer main and side chains.[12]

Amid the aforementioned plentiful research efforts being devoted worldwide to develop new membranes of lower cost and enhanced performance for acid and alkaline water electrolysis (and fuel cells),[7] nanocellulose membranes are particularly promising.[13] Originally employed as a filler able to enhance the water uptake, thickness swelling, and mechanical properties of PFSA-based membranes,[14] nanocellulose can also be employed by cross-linking it with 1,2,3,4-butanetetracarboxylic acid obtaining effective PEM free of polyfluoroalkyl substances (PFAS).[15]

Researchers based in different European countries are actively developing nanocellulose membranes as sustainable, low-cost alternatives to conventional fuel cell membranes for both anion- and proton-exchange membrane electrolyzers and fuel cells. Focusing on recent innovations, this study offers a European perspective on research on nanocellulose ion-exchange membranes for new-generation hydrogen fuel cells and water electrolyzers. Perspective is timely as research efforts conducted in Europe in the past decade (2015−2025) are approaching technology commercialization. This study highlights also how research policy promoting a cross-disciplinary and cooperative approach to research enabled the innovation behind the development of these enhanced membranes.

2. OBJECTIVES

Partly driven by the European Union's ambitious green hydrogen strategy outlined in mid 2020,[16] the key research objective of European research on nanocellulose membranes in fuel cells and electrolyzers is to replace expensive PFSA membranes with biobased, nonharmful, sustainable-by-design materials of superior performance and affordable cost. Superior performance translates into high ion conductivity, longer durability, high mechanical tensile strength (typically exceeding 50−90 MPa) to withstand operational pressures and swelling, and excellent gas barrier properties, crucial for electrochemical cell efficiency; while superior environmental performance is based on eliminated environmental and health issues associated with PFAS membranes. Fluoropolymers indeed are of high concern for environmental and human health due to their extreme persistence and high likelihood for human and animal exposure.[17]

Besides providing a highly attractive, environmentally friendly alternative to PFSA, the use of nanocellulose in commercial PEM and AEM membranes would provide European countries with increased energy security and autonomy. Nanocellulose, indeed, is sourced either from paper pulp or, even better, from agri-food byproducts so far chiefly dealt with as biowaste.[18]

European research efforts are aimed at enhancing the three main specific properties of nanocellulose for use in ion-exchange membranes, such as proton or hydroxide conductivity, mechanical strength, and water management without swelling. Due to plentiful hydroxyl groups of the cellobiose units comprising the cellulose polymer, pure nanocellulose in contact with water tends to rapidly swell and coordinate water molecules at the surface through extensive hydrogen bonding. Enhancing swelling in resistance and water management is thus critical for using these types of membranes employed as ion-exchange membranes or in the catalyst layer in both H_2 fuel cells and water electrolyzers. Swelling is generally suppressed by the material (composite) design and cross-linking via functionalization and composite (nanocellulose@polymer) membrane designs.[19]

Besides being a challenge, on the other hand, nanocellulose's hydrogen-bonding network is also an advantage because it offers a readily derivatizable molecular network to enhance proton and hydroxide ion mobility. In detail, to increase the inherently low conductivity of pure nanocellulose membranes (ca. 0.05 mS/cm), two successful strategies are (i to chemically derivatize nanocellulose, via the abundant hydroxyl groups in cellulose cellobiose units comprising the polymer, with ionic groups (carboxylates, sulfonates, quaternary ammonium groups, etc.) or (ii blending the bionanomaterial with ionic polymers, graphene oxide, carbon nanotubes, or metal−organic frameworks, thereby creating pathways for better ion transport.

Readers are referred to a recent review showing that nanocellulose-based membranes have reached even 130 mS/cm proton conductivity.[13] Typical objectives of EU-funded ongoing research projects on nanocellulose membranes for PEM fuel cell (PEMFC) and electrolyzers (PEMELs) are stacks targeting performances of >1.5 W/cm^2 at 0.65 V for PEMFC and 3.0 A/cm^2 at 1.8 V for PEMEL, with a degradation rate <5 μV/h (ensuring longevity over 1000 h).[20]

3. SELECTED ACHIEVEMENTS

In 2002, scholars in Sweden reported that cross-linking cellulose nanofiber (CNF) with 1,2,3,4-butanetetracarboxylic acid (BTCA) at an optimal rate of 20 wt % BTCA, a cross-linked membrane showing excellent ion conductivity (up to 8.1 mS/cm in 0.1 M KCl at pH 8.1, Figure 1) and selectivity was obtained.[15]

The green chemistry membrane preparation route adopted, furthermore, is readily scalable. BTCA is a biocompatible and

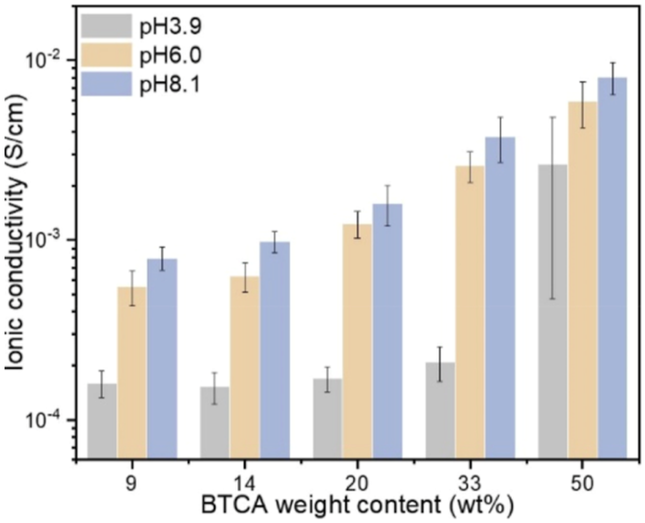


Figure 1. Ionic conductivity for cross-linked nanocellulose-BTCA membranes as a function of BTCA weight content at different pH. Reproduced with permission from ref 15, Copyright 2021 The Authors, CC BY-NC-ND 4.0 Creative Commons License.

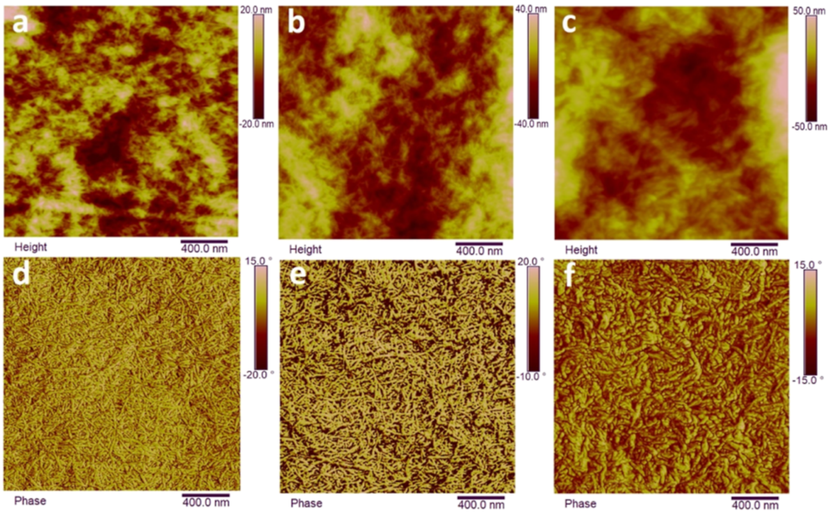


Figure 2. AFM topography and phase images of the cross-linked nanocellulose membranes with (a,d) 9 wt %, (b,e) 20 wt %, and (c,f) 50 wt % BTCA, respectively. Reproduced with permission from ref 15, Copyright 2021 The Authors, CC BY-NC-ND 4.0 Creative Commons License.

eco-friendly, formaldehyde-free cross-linker for cellulose, widely employed for optimal cotton textile application.[21]

In detail, CNF, glycerol, BTCA, and sodium hypophosphite (SHP) in solutions were mixed to achieve a CNF/glycerol solid weight ratio of 10:1 and a BTCA:SHP solid weight ratio of 1:1. The BTCA solid weight content was varied as 9, 14, 20, 33, and 50% with respect to the BTCA + CNF mass. The mixed solution was homogenized using a laboratory mixer for 5 min, followed by degassing for 1 h. The homogenized solution was poured in a plastic Petri dish and left in an oven at 60 °C overnight to dry. Cross-linked CNF membranes about 20 μm thick were readily obtained by removing the membranes formed in the Petri dish and heating the membrane for 15 min at 150 °C for cross-linking. After cross-linking, the membranes were immersed in excess water overnight, followed by rinsing three times to remove unreacted molecules.

The cross-linking not only improves the water stability of the nanocellulose membranes but also successfully introduces negative surface charge with higher charge density. Optimal ion selectivity was observed when the cross-linker is less than or equal to 20 wt % BTCA, due to formation of uniformly distributed nanochannels in the cross-linked membrane (Figure 2).

Highlighting the ease of membrane preparation, the team concluded that the discovery is promising for commercializing said nanocellulose-based membranes for PEM fuel cells and electrolyzers.[15] Indeed, a startup company was created in 2021 spun out from research at the KTH Royal Institute of Technology and Linköping's University, with the aim to commercialize this and related nanocellulose membranes replacing PFSA-based membranes with eco-friendly membranes of increased performance and lower cost.[22]

Similar results had been reported in 2020 by Lyth in Great Britain (and co-workers in Japan) at a meeting of the Electrochemical Society: cross-linking cellulose nanocrystals (CNCs) with sulfosuccinic acid (SSA) affords a PEM of improved ionic conductivity, enhanced mechanical strength, and reduced swelling in water.[23]

Two years later, the team reported that the conductivity of the CNC-35%-SSA membrane after boiling decreased by a factor of around six (Figure 3) compared with the initial conductivity due to de-esterification reaction and SSA leaching.[24] The researchers concluded that "it may be beneficial to explore different crosslinking reactions avoiding ester bonds", though the conductivity of the heated CNC-35%-SSA membrane was still significantly higher compared to the pristine CNC membrane.[24]

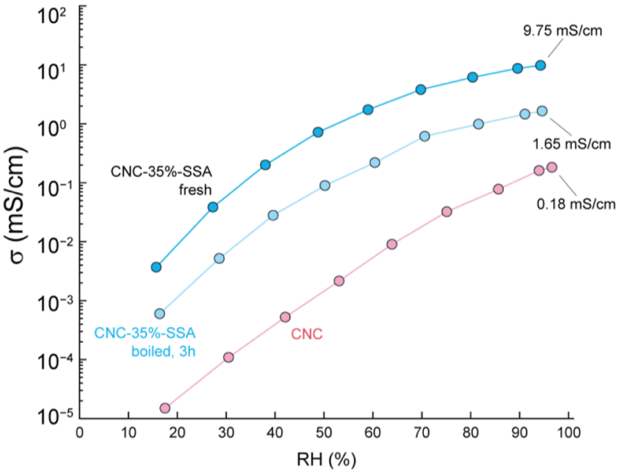


Figure 3. Through-plane proton conductivity of the CNC-35%-SSA membrane as prepared (blue) after 3 h of boiling in deionized water (light blue) compared to pristine CNC as a function of the relative humidity measured at 80 °C (red). Reproduced from ref 24, Copyright 2022 The Authors, CC-BY 4.0 Creative Commons License.

In 2025, a cross-disciplinary team led by Fontananova and Pagliaro in Italy reported the proof-of-concept study toward the development of technical and economically viable large-area anion exchange membranes (AEMs) combining high permselectivity with a low resistance to ion transport and stability during prolonged contact with a concentrated alkaline solution.[25] The latter are the conditions of alkaline water electrolysis. In detail, the researchers discovered that functionalization of polymerizable ionic liquid (PIL) (triethyl(4-vinylbenzyl)phosphonium tetrafluoroborate) with citrus nanocellulose CytroCell drives a substantial simultaneous enhancement of the chemical stability, ionic conductivity (Figure 4), and mechanical properties of the resulting composite membrane.

Sustainably sourced via hydrodynamic or acoustic cavitation of industrial citrus processing waste conducted in water only,[26]

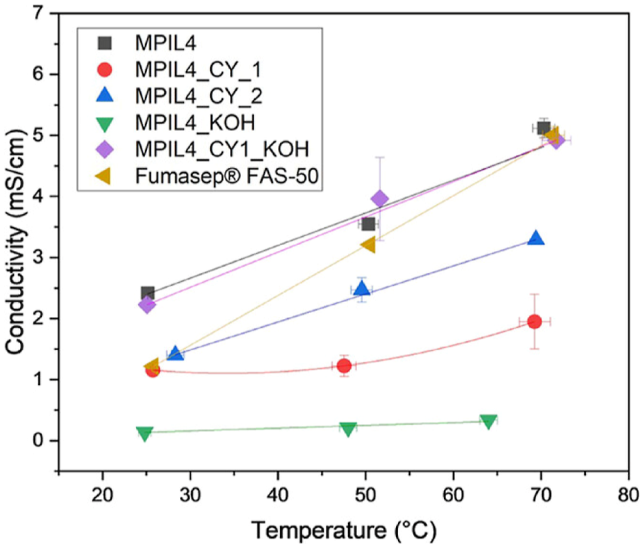


Figure 4. Conductivity vs temperature plots for the unmodified polymeric membrane (MPIL4), membranes modified with CytroCell (MPIL4_CY_1 and MPIL4_CY_2), and membrane MPIL4_CY_1 after treatment with KOH 5 wt % (MPIL4_CY_1_KOH). Reproduced from ref 25, Copyright 2025 The Authors, CC-BY 4.0 Creative Commons License.

CytroCell is a new family of nanocelluloses whose structure consists of submicron long cellulose nanofibrils negatively charged thanks to substantial (about 40%) esterification of the cellobiose units comprising the polymer chain with citrate groups.[27] The concomitant presence of citrate and hydroxyl groups provides CytroCell with an amphiphilic nature.

Molecular dynamics simulation studies and clustering phenomenon calculations indicate both excellent solubility in polar solvents including water along with strong chemical stabilization promoted by interaction of charged citrate groups.[27] This, *inter alia*, ensures quick dispersion of CytroCell in water where it forms a stable colloidal suspension thanks to a large negative zeta-potential (−25 mV).

Production of CytroCell is a direct process conducted in one pot that does not require neither chemical reactants to extract the cellulose nanofibrils nor energy-intensive homogenization to fibrillate the latter nanofibrils.[26] This is crucially important from the manufacturing viewpoint because the high capital and operating expenditure costs of nanocellulose production have limited widespread uptake in industrial applications for over three decades.[28] The aforementioned high operating and capital expenditure costs of conventional nanocellulose production are due to both the employment of harsh chemical conditions to extract and partly derivatize the cellulose nanofibers with anionic groups, followed by energy-intensive fibrillation.[29]

4. CROSS-DISCIPLINARY, COOPERATIVE APPROACH

It is instructive to review how rapid progress in the development of nanocellulose-based membranes for hydrogen fuel cells and water electrolyzers was actually achieved in Europe. After nearly two decades of failed attempts to commercialize nanocellulose-based technologies also in Scandinavian countries, Sweden's public innovation agency Vinnova in 2017 supported the creation of the Digital Cellulose Center.[30] Partners of newly created center of competence include academic institutions (RISE Research Institutes of Sweden, Linköping University, and Royal Institute of Technology) and private companies from the paper and pulp, packaging, electroactive materials, and energy storage sectors.

Its goals to 2027, namely, 10 years from inception, are to generate knowledge and competence relevant to industry by developing nanocellulose into electroactive functional nanomaterials for numerous applications, including printed electronic circuits and electroactive surfaces, so as to eventually make cellulose-based products "an integral part of a sustainable, digital society".[30]

The Center's achievements include ongoing commercialization of the aforementioned cross-linked nanocellulose membranes by partner company spun off from Linköping's University,[22] thanks also to close collaboration of academic researchers with industrial company partners of the Center.[31]

Besides offering the possibility to work together with companies that can quickly test, refine, and develop academic laboratory discoveries into viable products, this approach emphasizes the need to combine different disciplinary competences such as chemistry, physics, and engineering also in the host academic research groups involved in research.[30]

A similar approach was followed in Italy using funding from the European recovery program NextGenerationEU supporting cross-disciplinary research on green hydrogen.[32] Within a broad research project for development of new technologies for the hydrogen supply chain launched in 2022 involving numerous universities and research organizations, a specific line of activity dedicated to the development of materials and components not containing critical materials for anionic electrolyzers using AEMs was assigned to researchers from different institutes of the Italian Research Council.[33]

Ranging from membrane science and technology to catalysis, electrochemistry, materials science, and crystallography, the researchers involved in the research line had different competences and had never worked together before. The outcomes of their joint researches resulted not only in the development of the aforementioned new citrus nanocellulose-based membranes[25] but also in earth-abundant, high-performing Ni-based catalysts for AEM alkaline electrolysis.[34]

Clearly, room exists for integrating even more competence and skills in the development of low cost electrolyzers and fuel cells employing nanocellulose-based membranes. For example, artificial intelligence software is already used in AEM electrolyzers commercialized from Europe enabling further enhanced performance of the electrolyzer.[35]

5. CHALLENGES AND PERSPECTIVES

One key reason to develop low cost and eco-friendly nanocellulose membranes is to have an effective alternative technology to fluorinated polymer, that are PFASs posing serious health and environmental threats for which progressive ban started in EU countries already in 2023.[36]

Leveraging on nanocellulose's intrinsic properties like high gas barrier and robustness, ease of functionalization, lightweight, and outstanding health and environmental profile, current fundamental research efforts are focused on enhancing ion conductivity and chemical stability of the newly developed membranes. Simultaneously, to move from lab-scale innovation toward commercialization, applied research on manufacturing said membranes via green chemistry processes, preferably using roll-to-roll technology, is also being conducted in European and non-European countries. For example, researchers in Finland developed a roll-to-roll process for producing CNF coatings on paperboard as early as of 2016.[37] Showing evidence that similar research aimed at commercializing the roll-to-roll nanocellulose coating production technology is nowadays actively carried out in several non-European countries, scholars based in Taiwan lately reported a scalable process for fabrication of bacterial cellulose rolls.[38]

In brief, the technology hype cycle for nanocellulose is now over.[39] In Japan, by far, the world's leading countries in terms of nanocellulose manufacturing and industrial uptake in different products, at EXPO 2025, a paper and nanocellulose company producing both transparent and coarse phosphorylated CNF (P-CNF) slurries (Oji Paper), presented a nanocellulose composite proton-exchange membrane for hydrogen fuel cells as a sustainable alternative to perfluorosulfonic acid polymer membranes.[40]

Showing evidence that phosphorylated CNF membranes are fully comparable with state-of-the-art PFSA membranes under similar conditions (RH, temperature), in collaboration with researchers from the same company, the team of Masuhara had reported a maximum proton conductivity of 12×10^{-2} S cm^{-1}, equivalent to that of Nafion 212, measured at 20−80 °C and 95% RH.[41] Ascribed to the formation of a continuous proton conductive network consisting of closely packed phosphorylated cellulose nanofibers aligned with their long axis parallel to the membrane plane, the membrane has both high mechanical strength and high ion exchange capacity of 3.40 mmol g^{-1}.

This first example will likely be followed by nanocellulose and membrane manufacturing companies in Europe, North America, India, China, and other Southeast Asia countries. Nanocellulose production based on green and lower cost production routes using as a raw material, low cost and abundant agri-food processing waste will be shortly established in different world's countries, including European nations. The use of nanocellulose-based membranes in place of PFSA in PEM fuel cells is also advantageous in terms of enhanced durability under real fuel cell conditions because nanocellulose has high gas barrier properties, which decreases oxygen permeation through the PEM and thus reactive oxygen species (ROS) formation and membrane degradation.[42]

Today's alkaline water electrolyzers, in their turn, as separator anion-exchange membranes employ synthetic polymers with aromatic backbones covalently linked with ion-exchange groups including quaternary ammonium, piperidine, and imidazolium. Examples include zirconia/polysulfone composite containing 85 wt % filler (Zirfon)[43] or swelling-resistant polyphenylene sulfide derivatized with quaternary ammonium groups.[44]

Anion-exchange membranes made of pure nanocellulose have both limited conductivity and poor chemical stability in the highly alkaline (KOH/NaOH) electrolyte employed in AWE. On the other hand, it is enough to use a composite AEM made from polymerizable ionic liquid derivatized with phosphonium groups incorporating just 1 wt % CytroCell to observe remarkable stabilization of the membrane immersed in concentrated (30% KOH) alkaline solution (Figure 5).[25]

Whereas the unmodified polymeric membrane readily darkens in dilute and even more in concentrated KOH solution, the composite CytroCell@Polymer membrane retains its clear and transparent original appearance without any relevant change in the relative absorbance of the phosphonium group. Stabilization is due to the citric acid groups chemically bound to the CytroCell nanofibers, to which OH^- ions preferentially bind.

Future applied research studies will investigate the long-term stability using testing under accelerated aging conditions as well as state-of-the-art polymers commonly used as commercial AEM alkaline water electrolyzers. Finally, future chemical engineering research will address quality control in roll-to-roll processing, including integration into existing membrane electrode assembly (MEA) manufacturing lines for standardization and certification of nanocellulose-based membranes for both fuel cells and electrolyzers.

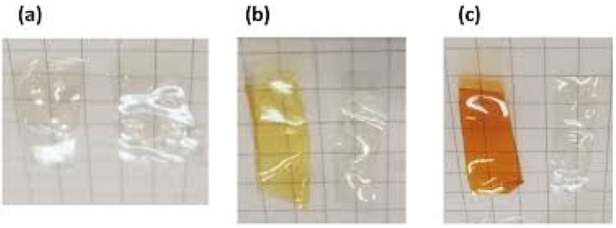


Figure 5. Image of the polymeric membrane MPIL4 (on the left in each photo) and the membrane modified with CytroCell MPIL4_CY_1 (on the right in each photo): (a) as prepared, (b) after immersion in KOH 5 wt %, and (c) KOH 30 wt % for 24 h. Reproduced from ref 25, Copyright 2025 The Authors, CC-BY 4.0 Creative Commons License.

6. CONCLUSIONS

In summary, contrary to what happened with the commercialization of Li-ion batteries, European academic and industrial research is at the forefront of developing innovative water electrolyzers and hydrogen fuel cells, including prototypes suitable for commercialization.

The study of how progress in the development of nanocellulose-based membranes for hydrogen fuel cells and water electrolyzers was actually achieved in Europe unveils that research policy efforts from national and EU funding bodies and programs were aimed at promoting systematic collaboration between academic research centers and companies as well as promoting a cross-disciplinary approach both to fundamental and applied research.

Research on nanocellulose membranes for hydrogen energy applications is flourishing also in China and in the USA (today's world's leading countries in nanocellulose research), Japan (world's leading country in nanocellulose manufacturing and industrial uptake), South Korea, and India. In common with research conducted in Japan in the field is the European focus on sustainability and on biobased sourcing of nanocellulose from abundant and readily available biological resources rather than employing harmful "forever chemicals" polymers such as PFSA. Newly developed national and European research policies in the field also borrowed from Japan the need to promote a cross-disciplinary and cooperative approach.

After more than two decades of failed attempts to commercialize nanocellulose-based technologies, the approach turned out to be successful. Nanocellulose-based membranes developed through research in academic and industrial laboratories in European countries are approaching commercialization in PEM fuel cells and are truly promising toward commercialization for AEM (and PEM) electrolyzers.

Likewise electricity, used for decades in all countries to power electric motors of all types, for cooling, lighting, and heating (via heat pumps), the same will occur with green hydrogen sourced from water via electrolysis and recombined with air's oxygen in fuel cells to generate electricity (and low-temperature heat) for both mobile and stationary applications.

With expanded nanocellulose production driven by new and growing demand from widely different industrial sectors, the membrane and hydrogen clean energy industries will likely become two key customers of the new nanocellulose industry emerging after the end of the technology hype cycle. In the process, European research on membrane technology for fuel cells and electrolyzers conducted between 2015 and 2025 has played a significant role.

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Notes

The authors declare no competing financial interest.

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Rosaria Ciriminna is a research director at Italy's National Research Council Institute of Nanostructured Materials. Developed in Palermo in co-operation with teams based in over 20 countries, her research work spans a wide field encompassing green chemistry, the bioeconomy, materials science, and biomedicine. Often cited for excellence in student mentoring, she has coauthored over 320 research papers and is one of Italy's most cited research chemists.

Daria Talarico is a research fellow at CNR-ITM. She obtained her PhD from the University of Rome Tor Vergata conducting work on electrochemical sensors. Her current research focuses on biobased and low-cost ion-exchange membranes for hydrogen fuel cells and water electrolyzers, including membranes functionalized with CytroCell nanocellulose both as anion- and cation-exchange membranes. Currently, she also teaches Chemistry in Italy's high school.

Enrica Fontananova is a research director at Italy's National Research Council based in Rende at the Institute of Membrane Technology. She graduated with honours in chemistry and obtained a PhD in Chemical and Materials Engineering in 2008 at the University of Calabria. Her main research interests include the development of membranes for different applications including ion exchange in electrochemical processes, desalination, and wastewater treatments. Her achievements are reported in over 110 research papers.

Mario Pagliaro is a research director at Italy's National Research Council based in Palermo. He has studied and worked in Italy, The Netherlands, Israel, and Germany. IntegroPectin, CuproGraf, CytroCell, NiGraf, AquaSun, AnchoisOil, LimoFish, and HyTan are some of the names he created to identify new functional materials and new enabling technologies jointly developed by his laboratory. Co-author of over 400 research papers and numerous books, in 2021, he was elected an ordinary member of the Academia Europaea.

ACKNOWLEDGMENTS

We thank Ministero dell'Ambiente e della Sicurezza Energetica under Piano Operativo della Ricerca "Ricerca e sviluppo sull'idrogeno" financially supported by the European Union—NextGenerationEU—M2C2 Investment 3.5, in the framework of the project PNRR Ricerca e Sviluppo sull'Idrogeno 2022-2025—Accordo di Programma "Idrogeno" (PRR.AP015.017.002), "Obiettivo 1—Produzione di idrogeno verde e pulito", LA 1.1.6 and 1.1.7. M.P. and R.C. also thank Ministero dell'Università e della Ricerca for funding under project "FutuRaw—Le materie prime del futuro da fonti non-critiche, residuali e rinnovabili", Fondo Ordinario Enti di Ricerca, 2022, CNR (CUP B53C23008390005).

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