Review and Perspectives on the Sustainability of Organic Aerogels

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Review and perspectives on the sustainability of organic aerogels

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TOC graphic

Abstract

Aerogels are exceptionally lightweight materials characterized by their high open porosity and remarkable specific surface area currently used across a wide array of industrial sectors, from construction to energy storage, and have great potential for expanding their applicability and unlocking new market opportunities. Driven by global economic growth and an intensifying environmental crisis, there is a growing demand for engineering innovations that prioritize sustainability. Aerogels are well-positioned to support these sustainability efforts. Their unique properties make them ideal for energy-saving solutions, environmental remediation, and more efficient use of resources. As the demand for eco-conscious technologies rises, aerogels are poised to contribute significantly to the development of greener, more efficient products and processes across multiple industries. The sustainability of aerogel technology is crucial for the mid-to-long-term future, yet its current status has been scarcely reviewed in the literature. This article explores and critically reviews significant advances on organic and hybrid aerogels in the current socioeconomic scenario with selected case studies endorsing their contribution to the UN Sustainable Development Goals. It also identifies research gaps while proposing innovative strategies to enhance the sustainability of aerogel production through the application of circular economy principles. Key strategies discussed involve the fabrication of aerogels using eco-friendly sources, such as biopolymers derived from biorefinery processes or from waste materials. Additionally, this article examines the development of methods for the reuse, recycling and end-of-life management of aerogels, along with the implementation of more efficient processing routes. Ultimately, this work highlights the need for comprehensive assessments of aerogel sustainability by through life cycle assessment (LCA), and evaluations of safety and toxicity. By addressing these critical aspects, the potential of aerogels to contribute to a more sustainable future appears highly favorable from both commercial and research perspectives, paving the way for a circular aerogel economy and providing a lasting impact to the society we live in.

Keywords: bio-aerogels; circular technologies; sustainable production; aerogel recycling; waste upcycling

1. Introduction

New production and consumption paradigms are emerging worldwide due to an overall expense increase derived from the scarcity of raw materials and low-priced energy.[1] Access to raw materials is of enormous importance for the economic stability of most countries, as they contribute to a robust industrial foundation, serve to produce daily goods, and are inextricably linked to the development of clean technologies.[2] However, especially after the COVID-19 crisis, global economic growth has resulted in an industrial bloom that accentuated the shortage of some resources, increasing industrial supply periods and prices with consequent economic inflation. As a result, recent international policies are addressing the identification of critical raw materials (CRMs) for multiple industrial sectors in Europe, the US, and Japan.[2–6] On the other hand, China is of significant geopolitical importance from mining and processing to the manufacture and trade of CRMs.[7]

The so-called *Twin Transition* envisions a carbon-neutral society by reinforcing digital breakthroughs and promoting green and sustainable technologies.[8] The significant material and energy reliance of Europe on third countries fostered the implementation of strategic projects for economic recovery and transformation (*Next Generation EU*). This enables exploitation of technologies and increases prospects for energy- and cost-efficient resources and process innovations.[2] Climate-neutral circular economy approaches are actively explored and implemented in all sectors and countries, with product reuse and recycling, as well as waste upcycling. The European Commission (EC) adopted the *New Circular Economy Action Plan* in 2020 to reduce strain on natural resources, ensure sustainable growth, and meet the EU's 2050 climate neutrality target.[9] The driving forces are the protection of the environment, the reduction of raw material dependence, and the sustainable boost of economic growth (create jobs, increase competitiveness, stimulate innovation, increase service life of products, improve quality of life in the long term). Similar initiatives have been launched by the USA (Inflation Reduction Act),[10] Japan (Green Innovation Fund),[11,12] and China.[13]

These new regulations, as well as market and societal demands, require the development of sustainable, innovative and advanced functional materials. A prominent class of materials that could address some of the mentioned challenges are aerogels, nanostructured materials endowed with unique properties e.g., high specific surface area (usually above 100 m^2/g), low bulk density (usually below 0.2 g/cm^3) and open porosity (usually higher than 85% with high presence of interconnected mesopores). Aerogels are especially attractive for a wide range of applications, from thermal insulation in buildings and industrial facilities,[14] to environmental (adsorbents for air, soils,[15] and water remediation,[16,17] selective binders for CRMs recovery,[18,19] sensors and catalysts,[20,21] sound absorbers[22]) and biomedical uses (scaffolds for regenerative medicine,[23] thermal insulators for photothermal oncotherapy,[24] dressings for wound healing,[25] drug carriers[26]). In addition, aerogels can be found in emerging applications in the food sector, where they may act as packaging materials with advanced functionalities (e.g., cushioning effect, thermal insulation, release or absorption of desired/undesired compounds) or food ingredients (e.g., fat replacers, delivery systems for active compounds, etc.).[27–29] The superior properties of aerogels in different fields resulted in a high scientific impact, such that the prestigious authority IUPAC (International Union of Pure and Applied Chemistry) identified aerogels as one of the Top Ten Emerging Technologies in Chemistry in December 2022.[30]

From an industrial point of view, the aerogel market is estimated at 1,155 million USD by 2025, with an average annual growth rate of 26% until 2030.[31] Advances in recent years encompass the use of various sources (inorganic materials, organic synthetic polymers, natural polymers, carbons, hybrid materials) for single component or composite aerogels, diverse morphologies (powder, beads, monoliths, mats, boards, films) and dimensions (from nanometers to meters), along with modelling, production scale-up, and health and safety assessments.

From a sustainability point of view, aerogel producers should decide on an energy-efficient drying process, probably the most critical step of the production line, with consideration on the material source and the intended use and performance. Furthermore, rational use of resources should include minimization of materials use, reuse of solvents, and recycling of unspent precursors, towards zero-waste in the production line. These must be also contemplated for the pretreatment of raw materials (e.g., extraction, derivatization, milling, purification) and the post-processing of aerogels (e.g., polishing, cutting, milling, carbonization, sterilization). Production costs can be optimized through the reduction of cycle time (for batch production), the increase of throughput (for continuous production), and the integration of processes. Studies on most of these technological aspects have been recently reviewed in the literature,[32,33] but life cycle and sustainability assessment studies are still scarce.[34,35].

While minimizing aerogel manufacturing time and costs, business strategies should also focus on sustaining and further extending the niche markets for this material. This perspective also addresses the circularity of different wastes that may feed the production of aerogels with an increased circular material use rate, reduced usage of raw materials, and lower energy consumption. For instance, developing more sustainable thermal insulation products could have important environmental and economic impacts as the building thermal insulation market is projected to almost double in the next ten years (from USD 25 billion in 2022 to USD 45 billion in 2032).[36] Similarly, the introduction of circular economy principles by using waste products in the wound healing domain will have an enormous economic impact, as the global wound care market is expected to expand to 28 billion USD by 2029. Similarly, aerogels intended as food grade oil structuring ingredients will position in the market of fat replacers, which reached 2.2 billion euros in 2022 in Europe and with an annual growth rate of *ca.* 6%.

The societal impact will be tied to both the lowering of the country's consumption footprint and its raw material self-sufficiency. However, sustainability and end-of-life management of aerogels, as well as rational use of resources for their production, have received little attention thus far, providing a technological and commercial challenge to the current state-of-the-art.

Sustainable aerogel production has recently evolved, with significant and exponential research growth rates.[37] The expected roadmap for this topic from different perspectives and approaches will be discussed in the following sections of this article: (i) identifying aerogels positioning in the United Nations (UN) Sustainability Development Goals (SDG) context (Section 2); (ii) performing a critical review of significant advances and searching for research gaps in innovative and (still) underexplored use of sustainable sources for aerogel production from biorefinery (Section 3), (iii) addressing wastes and by-products (Section 4); (iv) proposing various options for end-of-life of aerogel wastes by their recycling, reprocessing or upcycling (Section 5); and (v) proposing the implementation of process integration strategies and emerging technologies in aerogel production to minimize the consumption of resources and decrease energy use (Section 6). Finally, end remarks and other remaining challenges in the sustainable production of aerogels are compiled and emphasized (Section 7).

2. Aerogels in the UN SDGs context

The UN established 17 SDGs as part of the 2030 Agenda for Sustainable Development. To meet their requirements, the economic paradigm must change from linear to circular. This shift emphasizes responsible material usage and disposal, as well as recycling. Recycling has several advantages, including energy savings, natural resource preservation, and waste reduction. The management of solid waste has a significant impact on community health, as well as the natural environment.[38] Governments assign 4-10% of their total budgets to this duty as a first attempt to fulfil SDG13 (climate action).

Bio-based aerogels (or bio-aerogels) obtained from natural polymers are advantageous for their abundance, biocompatibility, and biodegradability.[28,29] Bio-aerogels are in harmony with the SDGs due to their inherent degradability, which ensures their disposal in accordance with the circular economy. Biocompatibility is another intrinsic feature of bio-aerogels that opens the sustainable application of these solutions in the biomedical, environmental, food, and packaging fields, aligning with SDG3 (good health and well-being) (Fig. 1). From one side, these eco-friendly solutions are sustainable alternatives to standard non-renewable medical or packaging materials, which are reducing environmental impact directly throughout their life cycle. On the other hand, biocompatibility makes bio-based aerogels food-grade materials, supporting their use as ingredients for sustainable and healthier diets. These sustainable applications of aerogels are being studied using different renewable sources such as chitosan, cellulose, poly(lactic acid), proteins, etc.[39,40] These derivatives could be obtained by the valorization of agricultural wastes (e.g., crustacean shell or sugarcane) or food industry by-products,[41] thus fulfilling the SDG12 (responsible consumption and production) and the SDG2 (food security, improved nutrition and promotion of sustainable agriculture). Additionally, aerogel sorbents manufactured by the valorization of agricultural and food wastes, or aerogels obtained from mineral phases (e.g., silica and clay), are sustainable alternatives with high capacity for water remediation and recovery of CRMs, such as Pt, Pd, Ag, Au, lanthanides or actinides from aqueous media.[42–44] In this regard, aerogels may contribute to SDG6 (clean water and sanitation) as well as to SDG14 (life below water).

Buildings and their facilities must have minimum energy requirements since the publication of the *2010/31/EU Energy Performance Building Directiv*e by the EU. Aerogels represent a new generation of thermal insulation materials with remarkable engineering applications due to their lightweight and extremely low thermal conductivity, as their nanostructure restricts the conductive heat transfer and the movement of gas molecules. Research and development on aerogels as new superinsulating materials is a way to commit to this policy with the ultimate goal of designing net-zero energy buildings. All these efforts are well linked with the SDG11 (sustainable cities and communities) (Fig. 1).[45] As an example, silica aerogels are extremely lightweight and highly effective thermal insulators with a thermal conductivity significantly lower than other commercial insulating solutions. This advantageous skeleton has fostered their use in green building construction or clothing, energy production, automotive, aerospace and military industries thus being in line with the SDG7 (affordable and clean energy) (Fig. 1).[46,47] Other applications of silica aerogels are being explored like their use as acoustic absorbers due to the low velocity of the sound transmission throughout the matrix of the aerogel thereby promoting the SDG9 (industry, innovation and infrastructure).[48] In these applications, research should focus on improving the mechanical, processability, and stability properties through the use of organic (polyurethane[49] or polyimide[50]), carbon[51] or hybrid (silica-organic[52]) aerogels, as they remain as the biggest challenges for aerogels efficiency and long-term performance.[53,54]

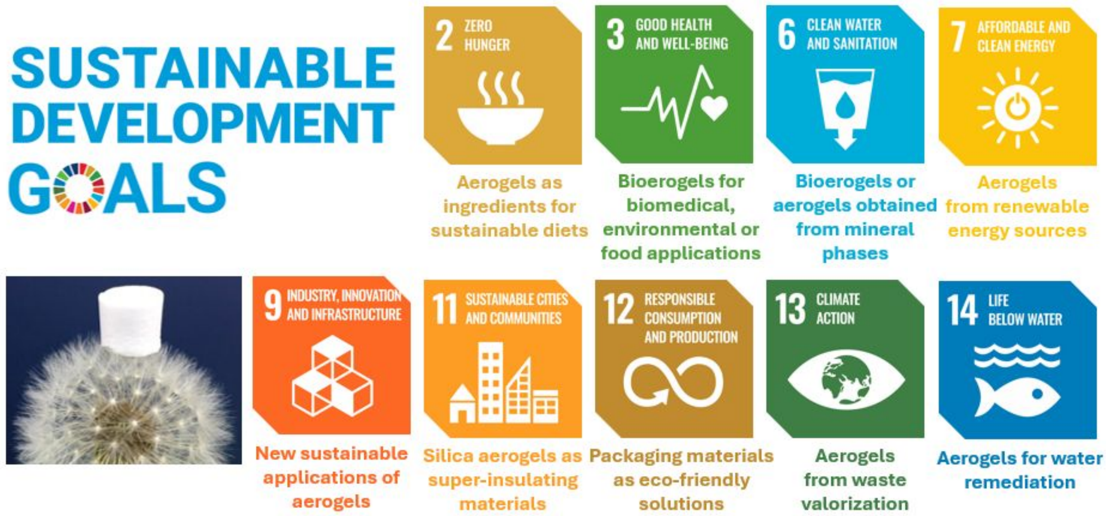


Figure 1. Outlook of the most remarkable contributions of aerogels to the UN sustainable development goals.

3. Aerogels from a biorefinery approach

Alternative sustainable sources for aerogel production are currently under evaluation. A key selection factor is the abundance of the source at an affordable cost to secure the supply. The biorefinery as a source of raw materials is especially attractive from an environmental standpoint (Figure 2). Indeed, biorefinery technologies allow for the efficient valorization, fractionation, and transformation of different biomass feedstocks in terms of mass and energy consumption.[55] Several economic and life cycle assessments available elsewhere strongly support the implementation of this biorefinery concept.[56–58] Conversely, this section focuses on the identification of research gaps.

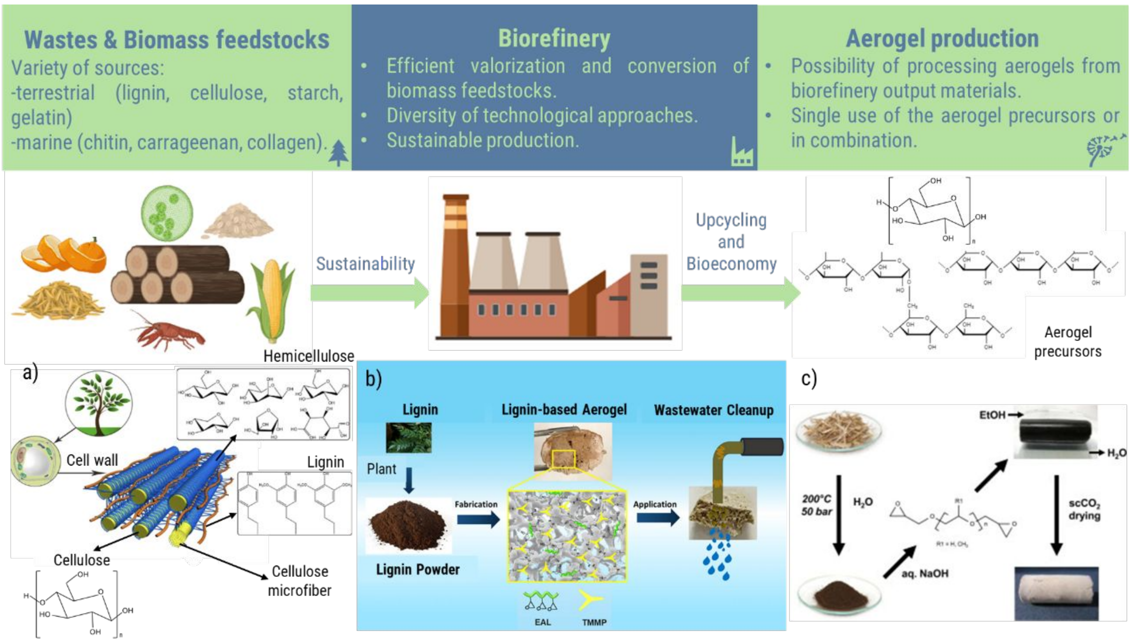


Figure 2. Implementation of biorefinery approaches in aerogel production: (a) Lignocellulosic biomass chemical composition.[55] Adapted with permission from ref.[59]. Copyright © 2017,Elsevier; (b) Lignin-based aerogel for wastewater remediation.[60] Adapted with permission from ref.[60]. Copyright ©2022, Elsevier; (c) Aerogel preparation from lignin derived from wheat straw.[61] Reprinted with permission from ref.[61]. Copyright ©2014, Elsevier.

Biorefineries employ several technological approaches, the most important of which depend on biomass feedstock attributes (e.g., origin and amount of residues) and product standards targets (yield, purity). In the bio-based economy paradigm, using these biorefinery output materials to make aerogels can contribute to upcycling into high added-value advanced materials. The sustainable production of materials from biomass should be supported not only by the source itself but also by using eco-friendly and safe production technologies with a low CO_2 footprint, avoiding the use of hazardous reagents, and preventing mass losses during the process cycle. Aerogel end-of-life and waste management should also be considered (cf. Section 5), as they should be safe for producers and customers, sustainable for the ecosphere, and economically feasible. The management of aerogel leftovers after use should be defined by design; otherwise, the sustainable production approach will be diluted or neglected.

Two main biorefinery sources can be used to produce aerogels (bio-aerogels): terrestrial and marine tissue wastes. The most common raw materials from terrestrial vegetal wastes are lignin, cellulose, pectin, and starch, while silk fibroin, gelatine, whey protein and keratin are from terrestrial animal wastes and by-products.[62–69] Agarose, chitin and derivatives (chitosan), carrageenan, and collagen and derivatives (gelatine) are the most used raw materials from marine wastes.[62,67,70,71] These raw materials are used alone or in combination with natural or synthetic components, for example, crosslinkers or other admixtures (e.g., nanoparticles), to tune the physicochemical properties of aerogels. It should be noted that the use of biorefinery-derived aerogels may be limited in some areas, such as in life science applications (biomedicine, pharmaceutics), due to the type, quality, and variability of the biorefinery source.[26,27] Finally, the choice of additives for the material design should be rational to avoid the underscoring of aerogel sustainable production.

4. Aerogels as a green way to waste upcycling

Economic development, population growth, and fast urbanization are associated with a significant increase in consumption and a consequent increase in waste. Currently, 2.1–2.3 billion tons of only municipal solid wastes per year are generated worldwide[72] and the global circular material use rate is generally low (e.g., only 11.5% in 2022 in EU-27)[73]. This translates not only in high managing and disposal costs but also in wasting of resources (land, water, and energy) necessary to produce goods.

Waste recycling refers to the reuse of existing waste material, which frequently results in lower-quality products with limited applications. The concept of waste upcycling refers to its reuse to fabricate upgraded or added-value materials, also known as waste valorization.[64] Aside from the transformation of the waste into relatively pure raw materials via biorefinery processes (cf. Section 3), waste upcycling can also be accomplished by simpler processing involving the integral conversion of the pure biomass into valuable derivatives.[74,75] Because no strong purification or extraction is performed in this situation, the high value of the resulting materials can only be achieved through complex supra- or macromolecular structures or architectures.[76,77] As previously mentioned, aerogels are regarded as high-value materials with outstanding textural properties. However, applications such as food and beverage (direct) packaging, cosmetics, or technical clothing are usually not considered for waste-derived aerogels,[78] due to a general lower purity than those obtained through biorefinery.

A first important step to allow for the usage of integral biomass as aerogel precursor would be waste upgrade from waste to by-product. According to the waste framework Directive (Directive 2008/98/CE), a waste ceases to be a waste and is classified as a by-product if specific conditions are met. For instance, the conversion of wasted biomass into food-grade aerogels would necessitate the establishment of a dedicated production chain, commencing with waste collection. The latter should adhere to rigorous food regulations to ensure the minimization of safety risks. In this context, it should also be pointed out that in most cases, biomass waste produced by food industries or consumers is currently classified as a by-product since it is utilized in biogas production, composting or animal feeding.[79] However, these strategies result in products of diminished value in comparison to those reaching the market. By contrast, the use of food by-products as aerogel precursors would allow their upcycling leading to high value materials.

Nguyen and coworkers conducted a thorough analysis of aerogels produced from waste materials,[38] focusing on the main outcomes of the research, such as aerogel characterization and performance. Conversely, in this section, an overview of the process will be provided, from the collection and processing method of the pure biomass into aerogels to their final performance and applicability of the resulting aerogels. The environmental impact of the whole process, from waste production to final reuse or disposal, will be discussed and research gaps and future directions will be identified.

4.1. Waste-based or byproduct-based aerogel preparation

Aerogels can be produced from waste or by-products that are dissolved or dispersed in a suitable medium, after chemical or physical treatment that will promote the formation of a 3D network. The mixture of textile fibers waste with a standard silica sol is an option to obtain composite aerogels with improved thermal and acoustic insulation properties, high water-contact angle and high flexibility.[80] Another example is the dissolution of recycled tire granulate rubber by and oxidant acid to form a rubber sol that can be easily mixed with the silica sol before its gelation. In this way, a hydrophobic efficient thermal insulator can be prepared, formed by a continuous rubber-silica aerogel matrix.[81] It can also be extended to compound other inorganic or organic sol-gel-derived phases. In some cases, phase separation and gelation can be induced by a non-solvent, as for example for aerogels prepared from cellulose-based textile.[82,83] Alternatively, waste-derived aerogels can be produced by direct mixing waste or by-product materials with EtOH or water/EtOH mixtures with increasing concentrations of EtOH[84] followed by supercritical drying. In this way, water originally present in the waste material is substituted with ethanol, which is then removed via supercritical drying. This process is particularly convenient when the waste material already presents a natural architecture (e.g., plant wastes and by-products) suitable for aerogel production.

4.2. Waste streams

Food and agriculture waste

According to the FAO 2024 "Global Facts" (FAO, 2024),[85] around 8-10% of global greenhouse gas emissions are associated with food that is not consumed with *ca.* 931 million tons lost in the supply chain, from after harvest, and prior to reaching retail shelves in 2021, and 1.05 billion tons of food wasted in households, food services and retail in 2022. This represents *ca*. 30% of all food produced worldwide for human consumption raising significant ethical concerns.

Food by-products are often represented by animal or plant tissues which are discarded during food processing such as swarfs, substandard materials or exhausted matrices e.g., from fruit juice and oil extraction. These cellular tissues can be treated as gel-like materials made by a complex biopolymer network, mainly structured by cell wall cellulose fibers, embedding water within intra- and inter-cellular spaces. The direct conversion of these "gel-like" materials into "aerogel-like" ones could thus represent a possible approach to sustainably turn critical biomass into shelf-stable ingredients without further generation of waste. For instance, by-products (external leaves and core) from industrial fresh-cut processing of iceberg salad were submitted to water-to-ethanol substitution and supercritical CO_2 drying, producing a white aerogel-like material which could be used as packaging, absorbent, or innovative carrier for both lipophilic and hydrophilic compounds.[84] When the same procedure was applied to homogenized substandard peas, colorless powders without vegetable sensory notes, but with high nutritional value and technological functionality were obtained.[86]

Food by-products have also been used to produce fully biodegradable pure[87] and hybrid[88] aerogels with high porosity. In the latter case, the food residue was homogenized and structured into a superimposed architecture by means of additional gelling agents (e.g., k-carrageenan, polyvinyl alcohol).[88] This approach allowed turning discarded salad leaves into aerogels for food applications. An important limitation of waste-derived natural polymers resides in their high hydrophilicity, which has derived into strategies for hydrophobic enhancement of the resulting aerogels.[89] Lignocellulosic aerogels were produced from spent ground coffee and apple pomace[90] with enhanced hydrophobicity through silanization in a liquid phase or by vapor deposition. Silanes are common hydrophobizing agents for cellulose, forming polysiloxane structures by reacting with the hydroxyl groups of the cellulose fibers through a condensation reaction.[91] However, it should be noted that Si-O-C bonds are easily hydrolysable in the presence of water.

Textile waste

Besides recent awareness about the use of fast fashion,[92] reality shows an increased consumption of textile products from 78 to 103 million tons in the last decade, and this tendency is still growing. Each European citizen discards an average of *ca.* 11 kg of textiles annually, most of which are disposed of in landfills or incinerated.[93] Landfill disposal has been forbidden in the European Union since 2016; incineration leads to the release of harmful chemicals and produces significant CO_2 emissions. The textile recycling process possesses a poor economic value, so research groups have explored engineering solutions to provide added value, such as using the fibers for mechanical reinforcement of aerogels.[80] In the case of cotton fibers, they can also provide a buffer effect for humidity regulation.

About 30% of textile are based on cellulose (cotton, viscose, Tencel). Till now, the main option of upcycling cellulose-based waste textile is dissolving the fabric and spinning fibers or casting films; this work is mainly performed on a laboratory scale or by small companies. Recently it was demonstrated that it is possible to make aerogels from cellulose-based textile waste.[82,83,94] Cellulose fabric, cotton or viscose was dissolved in ionic liquids, coagulated in water or in ethanol, and dried with supercritical CO_2. The bulk density was from 0.07 to 0.2 g/cm^3 and specific surface area from 300 to 400 m^2/g. These aerogels could be produced in the shape of monoliths or particles (Figure 3).

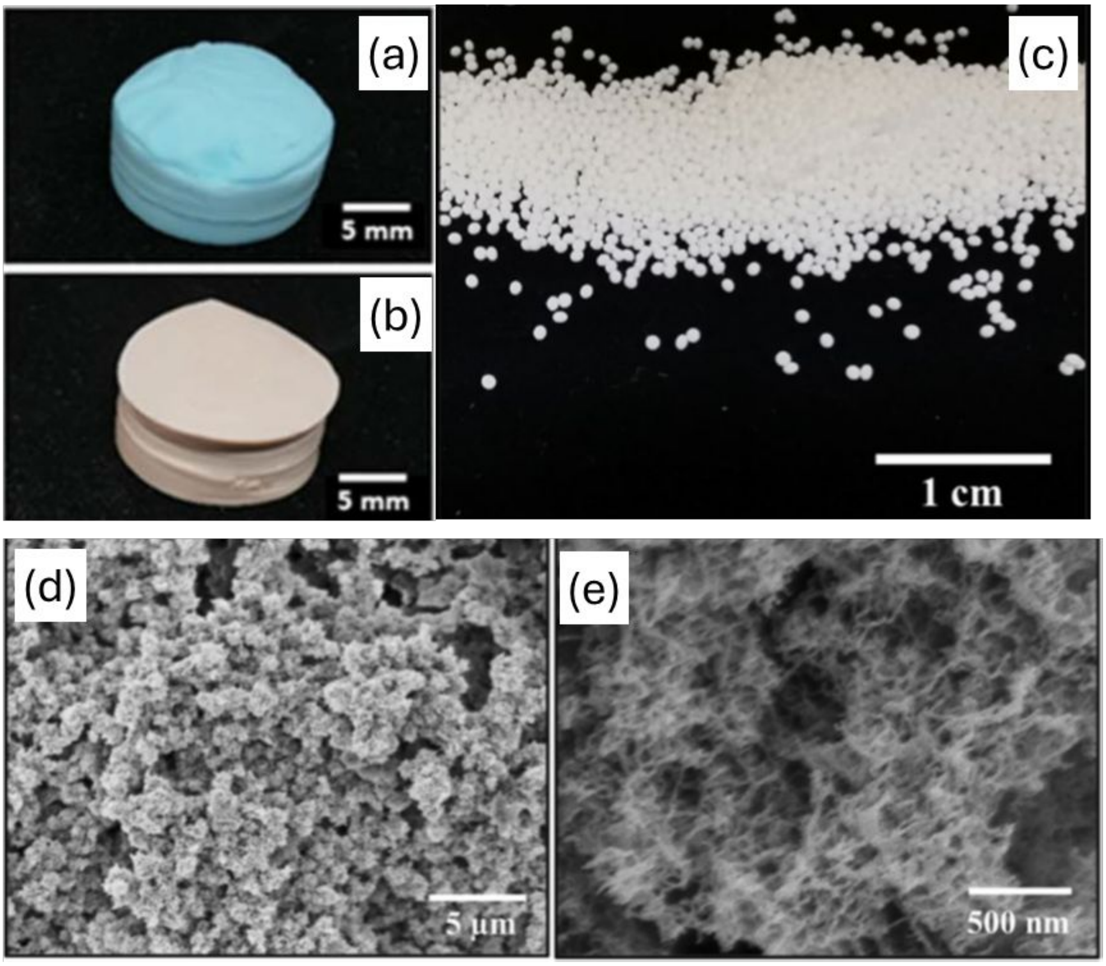


Figure 3. Aerogels prepared from waste textile ((a) rayon, (b,c) viscose), in the shape of monoliths[82] (bulk density 0.1 g/cm^3; specific surface area 330 m^2/g; porosity > 90%) and beads[83] (roundness 0.97 – 0.98, density 0.08 g/cm^3; specific surface area 400 m^2/g; porosity 97%); (d,e) their internal morphology, at different scales and similar for all aerogels, is imaged by scanning electron microscopy.[82,83] Adapted with permission from ref.[83]. Copyright © 2024, Springer Nature.

Paper waste

Paper is one of the most recycled municipal waste streams and *ca.* 72% of paper and pulp are produced from recycled sources.[95] However, the recycling process after several cycles causes fiber damage and reduced cellulose molecular weight.[96] The life cycle of this low-quality paper waste could be extended by its transformation into new and valuable aerogel-based products. Paper waste was used as a carbon source for the fabrication of carbon-based aerogels with outstanding performances as absorbents from water, with sorption rate values at least two orders of magnitude higher than those of activated carbon.[97] A typical preparation procedure consists in dissolving paper waste in water under stirring to obtain the pulp, drying of the fibers, and thermal treatment under inert atmosphere (including pyrolysis and chemical vapor deposition). These materials achieved exceptional surface area values (up to *ca.* 900 m^2/g) finding applications in the recovery of organic pollutants,[97] antibiotics, and oils/organic solvents from water[98].

Plastic waste

Around 400 million tons of plastic are produced annually worldwide.[99] Most plastic waste management strategies include direct disposal in landfills/the sea, and combustion, both of which have an important negative environmental impact. Polyethylene terephthalate (PET) is one of the most common plastic and is used in all sorts of consumer products due to its high stability and resistance to degradation.[100] This results in great interest in developing biodegradable alternatives that are stable enough for packaging applications. Besides the development of sustainable alternative materials, recycling of existing PET plastics is a major concern due to poor economic value and lack of environmentally friendly PET process. Recycled PET fibers can be potentially used to produce aerogels of high-value engineering applications (e.g., thermal insulation, CO_2 capture, and oil spill cleaning), but were only tested as PET cryogels so far.[100,101]

5. End-of-life of aerogel waste management by their recycling, reprocessing or upcycling

Reuse, reprocessing, repurposing and recycling are related concepts in the management of used materials, but with distinct differences. Reuse involves using an item again for the same or a similar purpose without significant modification. Reprocessing involves treating or processing materials to make them suitable for a new use. This often includes physical or chemical changes to return the material to a usable state. Repurposing is the act of using a product or material for a different purpose than it was originally intended, often with little or no alteration. Recycling involves converting waste materials into new materials or products, with the same application or not. All these concepts, especially the first three ones, are in many cases used indistinguishably, as very often reprocessing (also known as regeneration) is a prerequisite for reuse, which can also be considered as a similar, but more general, term to repurposing; recycling also requires reprocessing and/or implies reuse. The application of these concepts to aerogels up to now has been mainly devoted to recycling of the organic solvents used in the synthetic procedures or towards recycling of CO_2 used for supercritical drying of gels. However, recycling, reuse, reprocessing or repurposing of aerogels themselves has not been explored much yet.

The reuse or reprocessing of aerogels is an emerging research field for catalysis (i.e., reuse of aerogel catalysts) and environmental remediation (e.g., reuse of aerogel sorbents). It should be pointed out that when it comes to materials used as catalysts or sorbents, sustainability can be assessed based on several factors, including efficiency, longevity, and potential for reuse or recycling. In the context of a circular economy, sorbent materials that can be reused without significant loss of capacity are more sustainable because they can be used over multiple cycles, reducing the need for continuous production and disposal of new adsorbent materials. Although there are only a few such examples compared to the total number of aerogels explored for these specific applications, this can be attributed to the novelty of the concept rather than to the materials being unsuitable for reuse. The number of publications reporting reprocessing/reuse studies has recently increased significantly. This trend will reveal even more aerogel materials that can be reprocessed/reused.

In the field of catalysis, monolithic metal-doped carbon aerogels provide an example of easily reusable catalysts.[102] Carbon aerogels bearing metal nanoparticles dispersed homogeneously throughout their volume (Figure 4, M@C aerogels; M: Fe, Au, Pt, Pd, Ni, and Rh), prepared *via* pyrolysis of ferrocene-bearing polyamide aerogels and subsequent transmetallation, exhibited catalytic activity towards (a) oxidation of benzyl alcohol to benzaldehyde (Au@C or Pt@C), (b) reduction of nitrobenzene by hydrazine to aniline (Fe@C) and Heck coupling reactions of iodobenzene with styrene or butyl acrylate (Pd@C), with yields in the range of 85-98%. Due to their monolithic shape, all these catalysts could easily be removed from the reaction mixture. More importantly, these catalysts were reused five times just by transferring them into a new reaction mixture, without any need for reprocessing. The yields at the end of the fifth cycle were in the 70-86% range. Similarly, monolithic Cu@C aerogels, prepared *via* pyrolysis of Cu(II)-chitin aerogels, have been proven to be efficient chemoselective catalysts for the selective reduction of maleimides to succinimides under mild conditions, using hydrosilane as a hydrogen source.[103] Cu@C catalysts could be removed from the reaction mixture *via* filtration and then reused for at least six times without significant loss of activity.

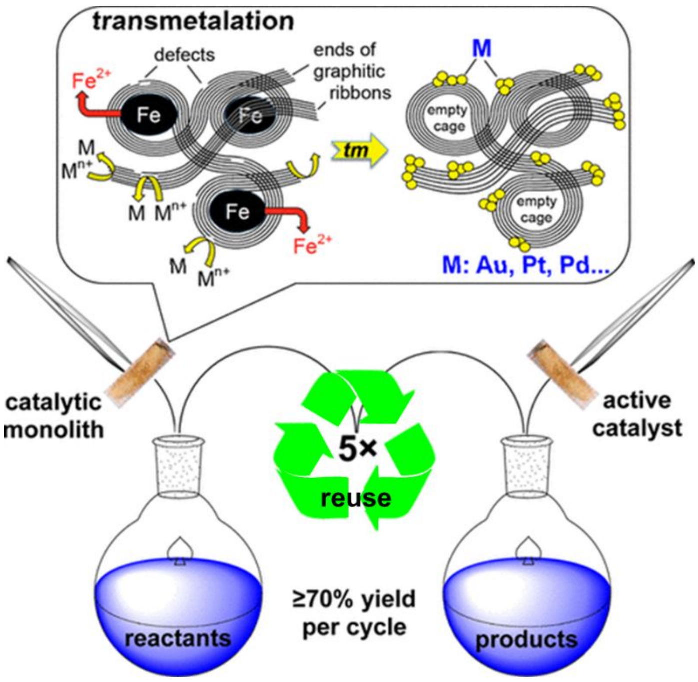


Figure 4. Transmetallation of Fe@C aerogels to M@C aerogels (M: Au, Pt, Pd, Ni, and Rh) leading to reusable catalysts. Adapted with permission from ref.[102]. Copyright © 2016, American Chemical Society. (The figure has been edited from its original version, where the authors had used the term "recycle"; in the context of the current understanding of the terminology, "recycle" is replaced with "reuse").

In the field of environmental remediation, more examples of reusable aerogels can be found. For these uses, reprocessing is also needed as the materials after adsorption/absorption of pollutants need to be stripped from the pollutant and regenerated before they can be used again. As demonstrated by the representative examples presented below, the regeneration process can be simple or quite tedious, depending on the materials and the specific application.

Crystalline imine covalent organic framework (COF) aerogels, prepared from the reaction of multifunctional amines with multifunctional aldehydes, were tested towards various environmental applications.[104] Depending on their chemical structure and pore size, COF aerogels could be used for the efficient removal of a range of organic solvents (both miscible and immiscible with water), organic dyes and inorganic micropollutants (gold nanoparticles) from water, and for the capture and retention of iodine vapor. COF aerogels with the best sorption capacities (16-35 times their own weight) for organic solvents, were solvent-exchanged with ethanol and dried again using supercritical CO_2. The reprocessed aerogels exhibited practically no loss of crystallinity or sorption capacity for 10 cycles. Similarly, the ones that showed the best removal efficiency (97%) for methylene blue were washed with acetone and methanol, and they were reused for four more cycles with practically the same performance in the removal of the dye.

Biocompatible biopolymer-based aerogels are by design suitable for environmental applications, as they have various potential coordination sites (e.g., $-COO^-$, $-OH$, $-NH$, $-NH_2$). One such example is polyurea-crosslinked alginate (X-alginate) aerogels, a new class of materials recently introduced,[105–107] which can be prepared from pre-formed alginate gels *via* reaction of the functional groups on the surface of the skeletal framework of the alginate (i.e., $-OH$) with multifunctional isocyanates, leading to the formation of a nano-thin polyurea coating over the entire alginate framework, which enhances the mechanical strength of the materials. Use of different multifunctional isocyanates allows for tuning of the material properties from the chemical composition perspective.[108] More specifically, X-alginate aerogels derived from the tris(4-isocyanatophenyl)methane (TIPM) are extremely stable in diverse aqueous environments (no swelling, shrinkage, or disintegration has been observed), including seawater. These materials can efficiently uptake organic pollutants (solvents,[109] organic dyes[110]) and inorganic pollutants, such as U(VI),[19,111] Pb(II),[109] Eu(III),[112] Th(IV),[112] Am(III)[111] and Hg(II)[110]. Their properties, combined with their high sorption capacity, allow for the reuse of these materials for several cycles. For example, in the case of Pb(II)[109] and Hg(II),[110] the materials can be treated with an aqueous solution of Na_2EDTA, washed with water and reused for at least three times (Figure 5a) without significant loss of performance. In the case of U(VI), which is the most extreme case, as X-alginate aerogels adsorb twice their mass (2 g g^{-1}), the materials can be reused for at least five times.[19] Similarly, silica–gelatine hybrid aerogels, prepared via co-gelation of gelatine and tetramethoxysilane, have shown high selectivity for the adsorption of aqueous Hg(II) in the presence of multiple competing ions, e.g., Cu(II), Cd(II), Co(II), Pb(II), Ni(II), Ag(I), and Zn(II) and can be reused for five times, after treatment with Na_2EDTA.[113]

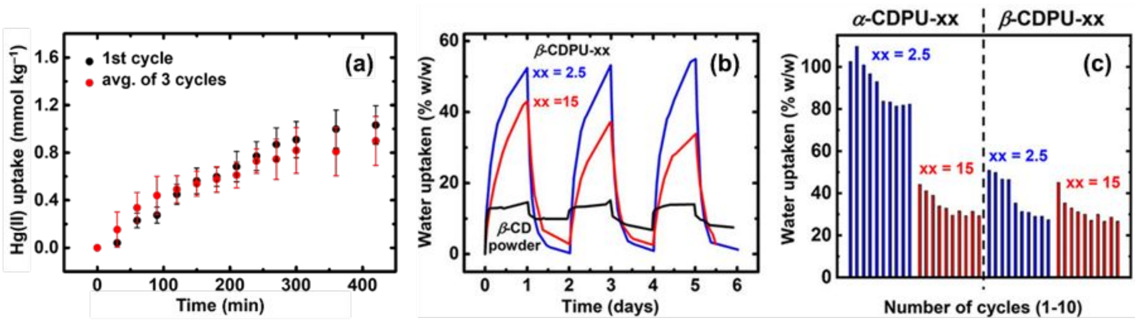


Figure 5. (a) Reusability of X-alginate aerogels for adsorption of Hg(II) ($C_{initial}$ = 100 ppb), after reprocessing that includes washing with aqueous solution of Na_2EDTA and water; (b) Three consecutive cycles of water vapor uptake between a high (99%) and a low (10%) relative humidity environment by β-CDPU-aerogels. (c) Ten consecutive cycles of water vapor uptake monitored every 24 h for α-CDPU- and β-CDPU-aerogels (xx: %w/w concentration of monomers in the sol). (b) and (c) Adapted with permission from ref.[114]. Copyright © 2019, American Chemical Society.

Another example of reusable aerogels with little treatment before being reused is α- or β-cyclodextrin-based polyurethane (CDPU) aerogels.[114] Upon exposure to a high-humidity (99%) environment at room temperature, these aerogels showed high water vapor absorption capacities (up to 108% w/w). These materials outperformed by far the corresponding cyclodextrins (in powder form) and the commercial products silica gel and Drierite™ (absorption capacities up to 20% w/w) (Figure 5b,c). Most importantly, owing to the balance of enthalpic and entropic factors, absorbed water could be released by just reducing the relative humidity of the environment to 10% at room temperature. CDPU aerogels can be reused for 4-5 times without any significant loss in performance, and they can be reused for at least another 5-6 times, as the water vapor uptake seems to have been stabilized, operating at 80% of their maximum performance. This facile regeneration is rather rare and practical and these materials could be used as desiccants in places where cold humid nights alternate with hot dry days.

Increasing interest in aerogel production and its applications has raised concern over their end-of-life management. High performance organic aerogels are typically composed of highly covalently cross-linked polymer networks, featuring pronounced chemical stability.[37] While this robust design provides organic aerogels with exceptional material properties, it makes them virtually non-recyclable, hindering their end-of-life management. When these aerogels reach the end of their service life cycle, they are either incinerated or disposed in landfills, leading to a loss in resources and a burden for the environment. Additionally, the economic loss of this linear produce-use-discard value chain is substantial and becomes especially important when the starting materials have high value. Reuse of aerogels reduces the burden of manufacturing energy; however, reapplying these materials eventually follows this linear economy model. Due to the lack of effective methods for recycling and valorization of the aerogel waste, valuable resources are lost, and the production of new aerogel materials continues to rely on fossil-based feedstocks and petrochemicals.

In stark contrast, the development of fully recyclable aerogels would provide the means for a sustainable circular economy. Therefore, various approaches are being explored to improve the recyclability of aerogels, specifically aiming to achieve closed-loop recycling. Recent ones focus on design of aerogel networks based on non-covalent interactions, including hydrogen bonding,[115] electrostatic interactions,[116,117] and metal coordination[118]. These non-covalent bonds can be easily broken and reformed under mild conditions, enabling the recycling and reprocessing of aerogels. However, while these reversible interactions offer a potential route to recyclable aerogels, their stability poses a significant challenge. The performance of these aerogels can degrade over time due to environmental factors such as moisture uptake or temperature fluctuations, which can weaken the non-covalent interactions and reduce the material's overall durability, raising questions about their viability for long-term applications.

A more promising strategy is the "design for recycling" approach which encompasses the introduction of reversible covalent linkages to fabricate organic aerogels that not only have excellent properties during their useful life but ensures their recyclability under selected conditions (Figure 6a).[119] The introduction of such bonds to the aerogel polymeric network facilitates the on demand depolymerization back into original monomers under energy efficient conditions. As the monomers are recovered in high purity and yields, they can immediately be reused to prepare fresh aerogels with identical properties as the original ones (Figure 6b,c).[120,121] This approach also allows partial depolymerization into soluble oligomers, that can promptly be used to prepare reformed aerogels.[122] Another key strategy is the incorporation of moieties into the aerogel structure that can selectively be cleaved under specific conditions into value-added chemicals and building blocks (Figure 6c).[121] The design of aerogel structures containing cleavable covalent bonds effectively addresses the environmental and economic challenges of traditional aerogels, paving the way for materials that combine excellent performance with the potential for sustainable, circular aerogel economy (Figure 6).

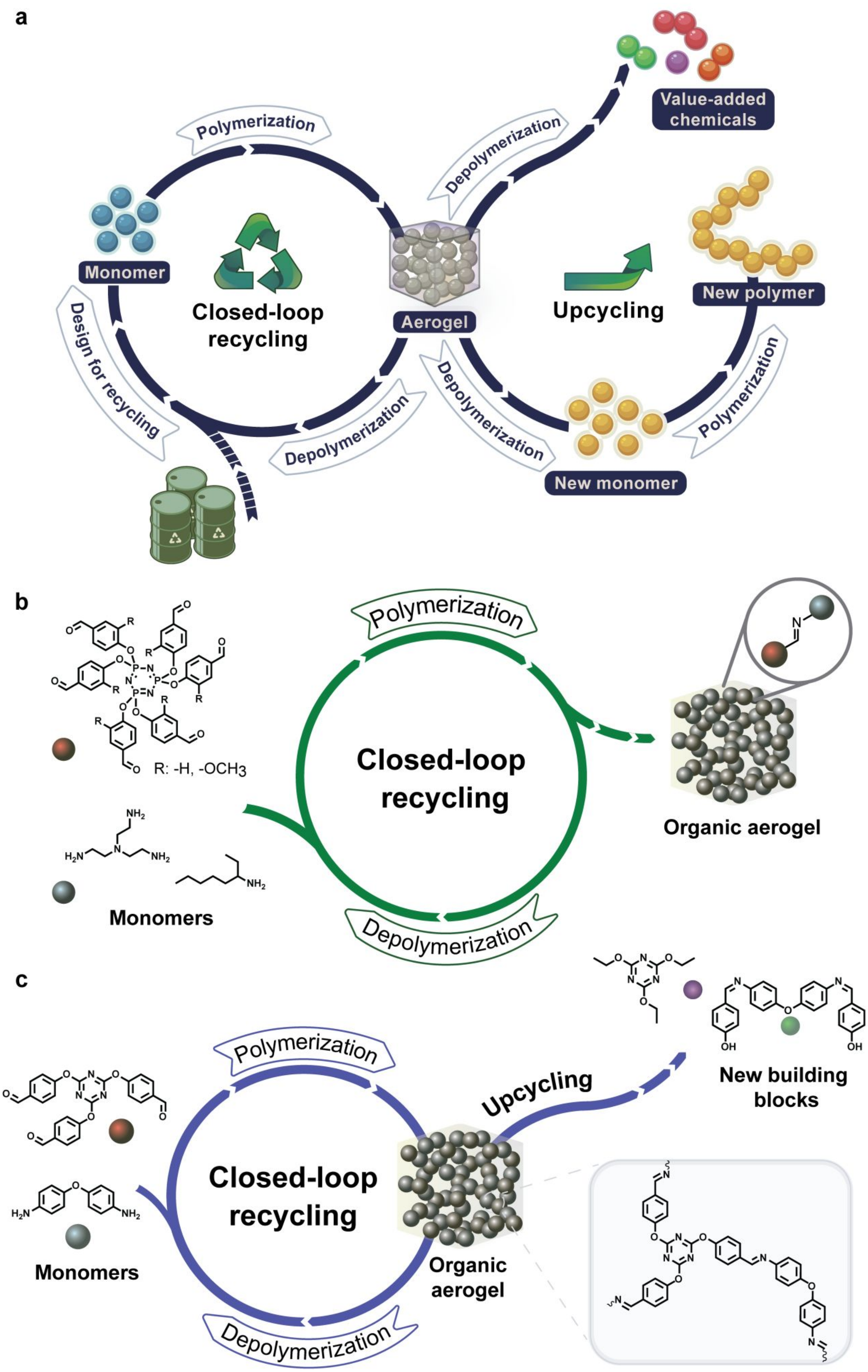

a
Polymerization
Depolymerization
Value-added chemicals
Monomer
Closed-loop recycling
Aerogel
Upcycling
New polymer
Design for recycling
Depolymerization
Depolymerization
Polymerization
New monomer
b
Polymerization
R: -H, -OCH3
Closed-loop recycling
Organic aerogel
Monomers
Depolymerization
c
Polymerization
Upcycling
New building blocks
Closed-loop recycling
Monomers
Organic aerogel
Depolymerization

Figure 6. (a) Illustration of the closed-loop recycling and upcycling scheme; (b) closed-loop recycling scheme for polyimine aerogels[120]; (c) closed-loop recycling and upcycling scheme for polyimine-cyanurate aerogels[121].

6. Process integration strategies in aerogel production

This section delves into process integration strategies in aerogel production (Section 6.1), focusing on adopting green and emerging technologies to develop bespoke solutions and expand application options (Section 6.2). Sustainable, high-performance, and personalized aerogels that meet specific user requirements can be developed through these innovative approaches, while minimizing resource use and environmental impact.

6.1. Evaluation of Process Integration Strategies in Aerogels Production

The integration of various production processes is crucial for increasing the efficiency and sustainability of aerogel manufacturing. Key strategies include hybrid sol-gel techniques, *in-situ* functionalization, and continuous flow processes.

The combination of standard sol-gel methods with advanced techniques such as supercritical drying can significantly reduce processing time and energy consumption.[123,124] The use of supercritical CO_2 for the drying phase in aerogel production is a highly efficient and environmentally friendly method. Supercritical drying technique not only reduces solvent residue in the final product to avoid the need for post-processing treatments, but also allows for the recycling of CO_2, minimizing greenhouse gas emissions. Most importantly, this hybrid approach not only preserves the structural integrity of aerogels but also enhances their physical properties. The solvent exchange in gel and supercritical drying steps combined in single apparatus can improve the efficiency of the aerogel production process.[124] Further on, solvent management should be considered. Instead of using pure solvents, technical solvent mixtures can be applied to improve the economics of the process. However, the effect of the solvent composition on the shrinkage processes and the duration of supercritical drying should carefully be evaluated.[125]

The integration of several processes into one, prompted further research on the topic of aerogel production. For instance, the processes of supercritical CO_2 drying and sterilization were integrated into a single one for the production of aerogels suitable for biomedical applications.[126,127] The integrated process produces decontaminated/sterile and ready-to-use aerogels while reducing processing time without compromising the aerogel's properties for regenerative medicine purposes.

Supercritical CO_2 can also be used for impregnation processes that enable the functionalization of aerogels, enhancing the material properties and application potential. *In situ* functionalization can be accomplished using co-precursor techniques or by incorporating functional additives into the sol-gel process.[128] Recently, the simultaneous starch aerogel formation and curcumin impregnation were reported to enhance curcumin's bioavailability and storage stability.[129] Functionalization of aerogels with natural bioactive components can also be performed after their production, for instance, via supercritical impregnation.[130] In addition to the impregnation of neat components, bioactive compounds found in plants can be extracted and impregnated in aerogels in situ using the integrated process of supercritical extraction-impregnation.[131] The resulting combination of two processes rendered savings in energy and processing time as well as minimization of extract loss and exposure to air and light. The impregnation processes can slightly change the morphology of aerogels (i.e., decrease specific surface area because of precipitation of compounds in pores), but the newly obtained functionalities of aerogels outbalance this disadvantage. Aerogels can also be used as superior carriers of hydrophobic synthetic drugs. The thus impregnated aerogels with beclomethasone dipropionate (a corticosteroid) show excellent aerodynamic properties at relevant doses, as confirmed by *in vitro* lung deposition tests, and the penetration into bronchial tissue as confirmed by *ex vivo* tests with porcine lung tissues.[132]

The shift from batch to continuous flow processes can streamline production, and reduce waste and energy usage. Continuous flow reactors enable precise control over reaction parameters, leading to uniform aerogel structures and improved scalability. A continuous-mode solvent exchange system can reduce the solvent consumption during the process to one-third with respect to the batch method.[133] In addition, the continuous supercritical drying process can efficiently produce aerogel particles.[134] A process design involving a counter-current extraction column with freely sedimenting alginate aerogel particles was proposed using a column of 1.0 m length. The drying of aerogel particles in a shorter column (0.5 m) could be achieved by increasing the CO_2 flow rate, resulting in a 20% reduction in the ethanol outflow mass fraction. Finally, the integration of energy recovery systems within the production process can capture and reuse wasted heat, enhancing overall energy efficiency. The integration of industrial waste heat recovery into smart energy systems represents a main opportunity to accomplish EU climate and energy objectives.[135] Such systems can particularly be effective in continuous flow processes where maintaining optimal reaction temperatures is critical. However, information on energy recovery system integration within the aerogels production process is still lacking.

6.2. Emerging Green Technologies for Aerogel Production

Emerging green technologies offer promising avenues for sustainable aerogel production. Innovative process strategies focus on the rational use of raw materials and energy, including 2D/3D-printing and plasma technology.

Aerogel materials are traditionally prepared using wet sol-gel chemistry, which involves sol preparation, sol-gel transition, post-treatment, and drying processes.[37] Alternative methods have recently emerged, such as the solid-phase route for perovskite oxide aerogels,[136] and the gas-phase route for carbon nanotube (CNT) aerogels[137]. These methods, whether traditional or newly developed, form the foundation for aerogel manufacturing, including both conventional and additive manufacturing techniques. Additive manufacturing, particularly 2D- and 3D-printing, emerged as a cost-effective production technology, suitable for both industrial scale-up and prototyping. This has positioned functional printing at the forefront of the material manufacturing revolution. Unlike traditional material removal, cutting, and assembly processes, printing is a "bottom-up" manufacturing method that builds materials from scratch. It is highly adaptable and potentially more cost-effective than traditional molding methods, allowing for the production of structurally complex parts that were previously unattainable.[138]

The first additive manufacturing of aerogels was reported in 2015, utilizing extrusion-based 3D printing of graphene oxide (GO) inks.[139] Since then, a wide variety of aerogels, including those based on graphene,[140] SiO_2,[141] resorcinol-formaldehyde (RF) polymer,[142] polyimide,[143,144] carbon,[145] cellulose,[146] metal,[147] semiconductor,[148] and g-C_3N_4[149] have been successfully printed. The technologies employed for printing aerogels include inkjet[150] and microvalve drop-on-demand printing on water-repellent surfaces[151] for microspheres, inkjet and screen for substrate-bound thin-films,[148] as well as microextrusion,[152,153] microgel-directed suspended printing[154] and the use of sacrificial templates[155] for creating 3D structures.

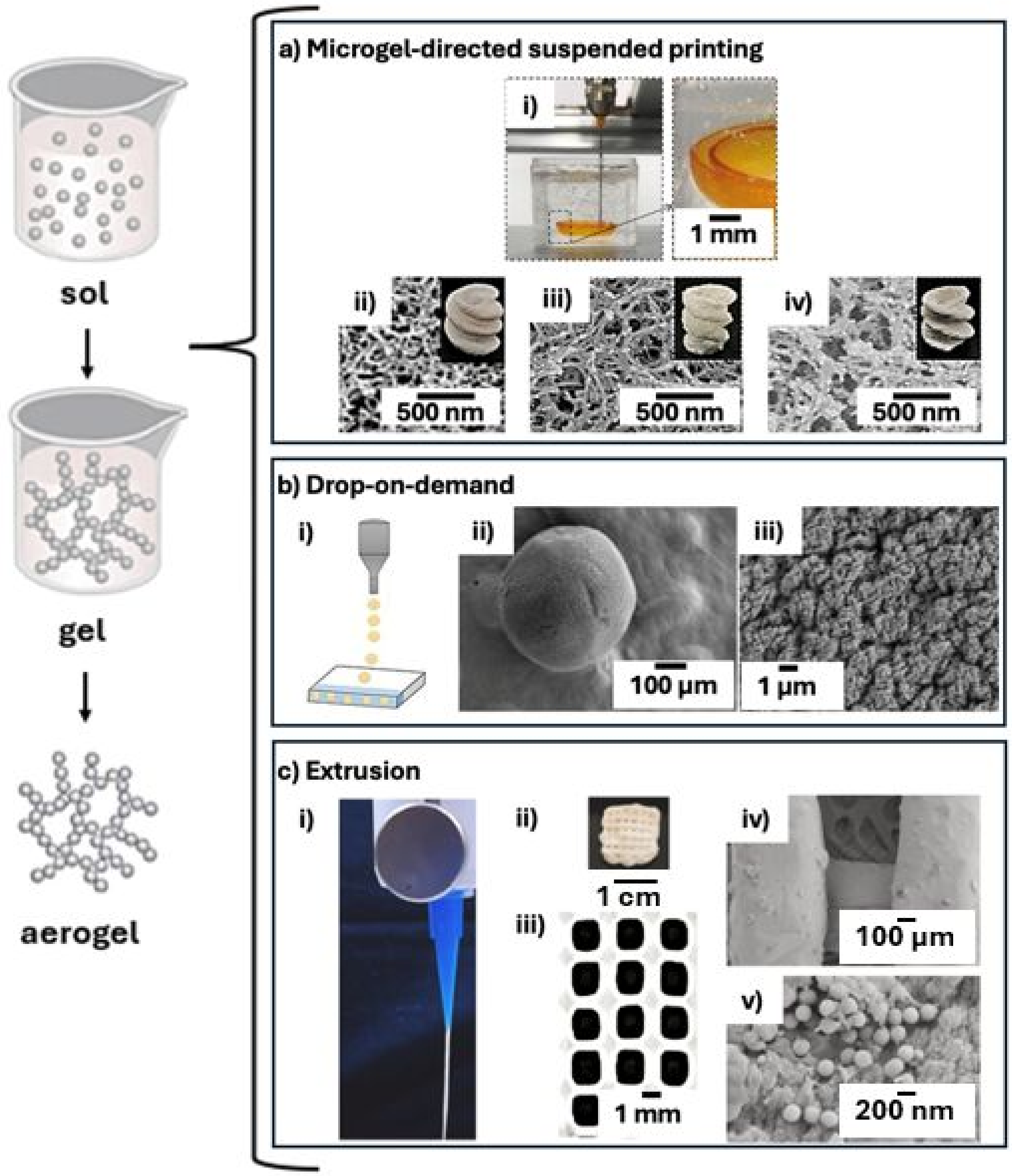


Figure 7: Conventional steps in the preparation of an aerogel (left). Integration of 3D-printing technology into aerogels production with selected examples (right). (a) Microgel-directed suspended printing set-up (i) Printing of a Kevlar nanofiber in a microgel matrix using such method,. (ii) cellulose, (iii) alginate and (iv) chitosan aerogels (and their corresponding SEM images) obtained by using the microgel-directed suspended printing.[154] Reprinted with permission from ref.[154]. Copyright © 2022, ACS Nano; (b) (i) Drop-on-demand printing process set-up on a superhydrophobic surface,(ii) low and (iii) high magnifications of SEM images of antibiotic-loaded alginate aerogels microspheres printed by drop on demand.[151] Reprinted with permission from ref.[151]. Copyright © 2022, MDPI AG; (c) (i) Extrusion-based 3D-printing set-up, (ii) Visual appearance of 3D-printed alginate aerogels, (iii) 3D-pattern observed on the hydrogel-based scaffolds. SEM images at (iv) low and (v) high magnifications of upconversion nanoparticles decorated alginate aerogels.[153]. Reprinted with permission from ref.[153]. Copyright © 2024, Elsevier.

Despite the fabrication method, aerogels are generally fragile due to their low solid content and nanoscale skeletal architecture. However, compared to traditional aerogel shaping methods, "bottom-up" additive manufacturing through functional printing offers unique advantages. In many cases, these methods allow for the design and shaping of aerogels on a microscale, addressing a significant limitation of traditional sol-gel and mechanical processing methods. The ability to control nanostructures through aerogel chemistry, combined with the macrostructural precision enabled by various printing methods, allows for adaptability and precise control over aerogel designs from nanometer to centimeter scales. This approach significantly shortens, simplifies, and improves the processes from basic chemicals or nanomaterials to functional device components, while also reducing production costs. Additionally, additive manufacturing facilitates the assembly of multi-materials with different functions and structures within different regions of an object,[144] a capability that is typically not possible with traditional fabrication methods (Figure 7). This customization allows to produce aerogels tailored to specific applications, minimizing material waste and enhancing performance. Recently, there has been a growing trend toward using bio-based precursors in 3D printing of aerogels, such as cellulose,[156] alginate,[157,158] silk fibroin,[159] further enhancing the sustainability of aerogel production in fields like food and medical industries.

Plasma technology is another green alternative for aerogel synthesis and surface modification, producing aerogels with unique functionalities. Plasma discharge can facilitate the formation of porous structures and enhance the material surface properties without the need for harsh chemicals. Plasma treatment can be also a fast and versatile technique for deposition of protective hydrophobic and oleophobic polymer layers on hydrophilic biopolymer aerogels. For instance, hydrophobic modification of biopolymer aerogels (derived from alginate, cellulose, whey protein isolate, and potato protein isolate) was performed by cold plasma coating.[160] While the porous structure of aerogels stayed intact during the plasma treatment, polymerization inside the aerogels pore led to the generation of new porous moieties and resulted in a significant increase in the specific surface area.

7. Current consideration on LCA of organic aerogels

LCA is the systematic mapping and evaluation of energy and material flows to critically assess the sustainability of a material or process across the entire life cycle. LCA analyzes the environmental impacts of resource consumption, material production, by-products, waste, and emissions. LCA evaluates a specific design or process in a "cradle-to-gate" or "cradle-to-grave" approach. Obviously, such an analysis is more straightforward for materials that have been applied, or are close to being applied, at an industrial scale. For materials that are still at the laboratory or small pilot scale, many assumptions are required which limit the confidence in the analysis.[35]

According to a 2021 survey of market data,[161] over 98% of the aerogel market is composed of silica aerogels, used predominantly for industrial, pipeline and battery thermal insulation and/or protection. Hence, the LCA of silica aerogel production through different synthesis routes has been evaluated, albeit in different frameworks and using different methodologies.[162] Thus, even for silica aerogels, it is therefore not possible to come up with a single measure of environmental impact. However, it is clear that for silica aerogels, the raw materials account for a significant fraction of the embodied emissions.

The high impact of raw materials for silica aerogel production made the development of more sustainable, non-toxic, yet cost-effective raw materials a key target of the aerogel scientific community and, to some extent, the aerogel industry. As emphasized throughout this perspective article, the search for sustainability is a key driver for bio-based aerogels, with commonly studied biomass raw materials including biopolymers such as cellulose, alginate, starch, chitosan, gelatin, and whey protein.[25,27,63,71] However, no bioaerogel products are currently available on the market at industrial scale production volumes, and there are no large-scale production facilities capable of manufacturing bioaerogels in sufficient quantities to meet real-world applications. Consequently, there is an urgent need to consider the LCA of aerogels, as these materials constitute an environmental impact due to their potential production requirements and the existing LCA studies on bioaerogel production are still limited to laboratory-scale analyses. These studies primarily focus on the "cradle-to-gate" scope, sometimes neglecting downstream processes such as utilization and end-of-life (EoL) stages (i.e. product disposal, solvent use and recycling, chemical recycling, and energy consumption).[35,163–168]

Although we are not yet able to evaluate the real industrial LCA of biopolymer aerogels, certain aspects can be predicted based on previous studies with biomass processing with other technologies, at small scale for these aerogels or with silica aerogel in industrial production. These studies remain a critical guide for the future development of biopolymer aerogels. Although biopolymer raw materials are inherently more sustainable, extraction methods, modifications, compounding processes, and drying methods can significantly influence their overall environmental impact. As an example, corn cultivation to obtain starch causes a high marine eutrophication (MEP) (54.9%), a high terrestrial ecotoxicity (ET) (50.5%) and a high land occupation (40%).[35] Furthermore, although the most part of the needed chemicals during the fabrication of aerogels do not remain in the end product, the way in which they are handled is critical for minimizing environmental effects, especially during the stages of solvent exchange and aging steps, where large amounts of solvents are used. Ethanol is one of the most utilized solvents and therefore, it can represent *ca.* 50% of the global warming potential (GWP) in all types of aerogels and is one of the main contributors to abiotic depletion potential (ADP) and photochemical oxidation.[169] Methanol is also commonly used, and by reducing the number of solvent exchanges carbon emissions can be reduced by up to sevenfold.[170] The drying process, in particular, remains a major challenge for LCA evaluations of aerogels because its significant energy and solvent demands strongly depend on the specific implementation, e.g., on the detailed equipment and process engineering. LCA of the drying step in commercial silica aerogel production provides insights for future optimizations in other aerogel sources. The drying process is both a challenge and a defining feature for aerogels, as it significantly influences their properties and environmental impact. Different drying methods have varying environmental impacts. Freeze drying, commonly used for biopolymer aerogels, is known for its very high energy consumption.[162,168,171] Ambient pressure drying and supercritical drying involve substantial solvent use due to the close correlation between drying processes and solvent types, leading to both high energy consumption and environmental concerns.[172,173] Recent studies have highlighted the need for enhancing the sustainability of the production through processes that minimize, eliminate, or recycle solvents and CO_2 (*ca.* 95% recycling rates can be obtained) without compromising aerogel properties.[35,166,174] Furthermore, optimizing energy consumption in fabrication processes and utilizing renewable energy sources can greatly reduce the carbon footprint.

The environmental repercussions of aerogels can be evaluated through six parameters: GWP, acidification potential (AP), ADP, eutrophication potential (EP), ozone depletion potential (ODP) and photochemical ozone creation potential (POCP).[175] The transition from lab-scale aerogels to pilot and industrial scales would reduce in a big proportion the environmental impacts. For example, in the scale-up production of starch aerogels from lab-scale to pilot, the GWP and AP reductions would be of 72%, while from lab to industrial it would be of 95%.[166] The reductions in EP would be of 61 and 93% for pilot and industrial, respectively; for ODP of 81 and 96%; the electricity use would be reduced in 89 and 99% and the primary energy demand (PED) in a 74 and 95%.

Aerogel waste management introduces further uncertainties in LCA. Biopolymer aerogels, primarily composed of polysaccharides or proteins, are generally considered non-toxic and biodegradable. However, their disposal can be complicated by the inclusion of inorganic or organic crosslinking agents (e.g., chitosan) or surface modifications (e.g., hydrophobization).

Composite structures further add to this complexity, and there is currently a lack of comprehensive information regarding their disposal methods and long-term environmental impacts. Finally, tailored LCAs are required for different application scenarios, from thermal insulation to environmental remediation and biomedicine, each with distinct environmental impacts. Addressing these challenges will be crucial for advancing biopolymer aerogels toward sustainable large-scale production and real-world applications.

8. Conclusions, future directions and final remarks

Despite the efforts to revalue waste materials towards innovative uses, aerogels produced directly from waste is still in its early stages. It is important that circularity not only focuses on waste recycling and reuse, but also considers that the processes for aerogels manufacturing or post-modification must be sustainable and scalable, with products being ideally reusable or recyclable. LCA for the production at all levels (from waste collection to end-of-life disposal/reuse of the developed materials) must be done by an economic analysis of the environmental impact considering the entire process.[176] Economic viability cannot be ensured solely by defining aerogels as high-value materials, but specific indicators (e.g., financial rate of return to companies and society, benefit-cost ratio) should be evaluated. Also, the alignment with specific United Nations SDGs should be reviewed.[177] In summary, there is still a research gap on the sustainable production of aerogels, which will motivate researchers to develop future aerogel-based materials.

Aerogels based on natural resources/waste fractions have some common challenges related to the large variations in the quality and composition of the raw materials. Even when the same biomass is used, this does not guarantee consistent biomass composition, as it is susceptible to significant variations depending on various factors, such as the climate history. Consequently, the properties of the fractions and final products (e.g., color, mechanical and chemical stability) may vary, potentially affecting the process. As an example, impurities originating from plant proteins may lead to catalysts deactivation during initial biomass processing.[178] Consequently, the aerogel production process must be continuously adjusted to accommodate the varying raw material quality, which is currently not considered in process analysis and LCA. In order to tackle this problem, stronger involvement of high-resolution climate modelling in the planning of material production processes is required. Recently, the term "climate-informed engineering" was suggested to address this issue[179] by training a new generation of engineers, who consider climate information in their engineering services similar to the way the economic aspects are considered in their products. Given that bio-based aerogel production combines numerous aspects of material science, engineering and chemistry relevant all over the world, it can serve as a demonstration field for this timely initiative and thus also improve the process sustainability.

On the other hand, the evaluation of the human and environmental impact will help to advance in the development and improvement of aerogels to address new health and environmental challenges within the context of the circular economy while complying with the *One Health* concept established by the World Health Organization.[180] The ecotoxicity and health risk assessment of aerogels are not specifically regulated, as aerogels do not require registration as nanoforms. Nevertheless, their nanostructured design can raise concerns about a possible hazard assessment, which needs to be addressed. For regulatory purposes, several aspects needed to be considered. As the toxicity inherent to aerogel exposure is not expected in general, the bioactivity of inhalable or ingestible fragments due to their high inner surface area can raise concerns.[181] The pulmonary route due to inhalation of aerogel nanoparticles as material dust and the consequent pulmonary deposition[182] is the main route of human exposure to the eventual toxicity of aerogels, since fine particles with diameter smaller than 2.5 µm penetrate the alveoli and even reach the cardiovascular system when smaller than 0.1 µm. Professional exposure during the installation and removal activities of insulation materials is one of the most frequent applications where humans are exposed. The use of aerogels on an industrial scale requires the implementation of safety regulations for workers involved in their production and exposure to these nanostructures. Moreover, these particles can be dispersed into the surrounding environment and can circulate in air, soil, and water. Thus, global regulation of their ecotoxicity is needed to prevent any risks to the health of the biosphere and to limit their associated pollution.[183,184] Additionally, appropriate safety regulations are involved in the manufacturing of aerogel products, identifying hazards for the environment, and human and animal health.[185,186] Since aerogels can comprise several chemical compounds, the composition of each ingredient should be classified as “non-hazardous” in order for the aerogel itself to be considered non-hazardous.[187]

There are still insufficient studies on the potential toxicity of certain aerogels. A couple of years ago, a systematic toxicological workflow, used to test nanomaterials, was proposed to evaluate and classify aerogels based on their safety profile.[182] Nineteen aerogels, both organic and inorganic, were compared using a 3 Tier evaluation. The materials’ biosolubility and oxidative potential were tested in Tier 1, the material's toxicity in alveolar macrophages *in vitro* in Tier 2 and intratracheal instillation in Wistar rats in Tier 3. All aerogels showed good biocompatibility, except for the case of polyurethane aerogels where a low toxicity potential was detected in Tier 2. From all tested aerogels, only a moderate, transient and reversible inflammation in the lung was found for polyurethane aerogels in Tier 3. In addition to these basic toxicity tests, it is mandatory to evaluate long-term exposition, repeated administration evaluation, bio-absorption, distribution, metabolism and excretion to ensure safe use in biomedical applications.[188,189]

From a data mining perspective, leveraging AI and machine learning algorithms can optimize production parameters, predict process outcomes, and reduce resource consumption. AI-driven models can identify the most efficient pathways for aerogel synthesis, minimizing the use of raw materials and energy. This direction has been successfully used for aerogel production and application optimization. For instance, it was shown that deep reinforcement learning (DRL) with the diffusion-limited cluster–cluster aggregation (DLCA) algorithm can be applied for microstructural optimization of silica aerogels.[190] Machine learning-based multi-objective optimization using NSGA-II-study of modelling has been applied for an aerogel glazing system in the subtropical climate in order to minimize the total heat gain and maximize the indoor illuminance transmitted through the system.[191] Moreover, an artificial neural network (ANN) was developed for predicting the fractal properties of silica aerogels, given the input parameters for a DLCA algorithm. This approach of machine learning replaces the necessity of first generating the DLCA structures and then simulating and characterizing their fractal properties.[191,192] Collaborative robotics were also tested in combination with machine learning tools to accelerate the design of conductive aerogels from mixtures of $Ti_3C_2T_x$ MXene, cellulose and gelatine with programmable properties.[193]

The visibility of aerogel-based materials and their impact as a top emerging technology have dramatically increased in the last decade. Overall, organic aerogels are multi-functional materials that can be obtained from synthetic or natural materials and have a wide range of applications. The industrial sectors for waste collection (food, textiles, cosmetics, cattle), biorefinery (lignocellulose production), and applications (construction, biomedicine, pollution remediation, critical raw materials recovery, food) are the target groups for expanding the sustainable production and use of aerogels. The awareness and possibilities of these advanced materials can be boosted by new and ongoing international initiatives among the aerogel scientific community. These initiatives include the implementation of an international association on aerogels looking for a harmonized voice for the scientific community, the general public and other stakeholders; and to increase networking, training, and other outreach activities. A redefinition of the aerogel term by the IUPAC Association was urgent and has been recently deployed to meet a consensus aiming at limiting the array of materials falling within this material category.

The snapshot on aerogel technology provided in this perspective article unveils a promising present and near-future market outlooks alongside an active and growing scientific community. The glimpse of mid-to-long-term future trends on aerogels technology and applications herein provided is optimistic with novel uses and fast-growing market shares. It also highlights prominent research to adapt conventional aerogel production towards the paradigms of circular economy, and raw material and energy efficiencies. The sustainability principle must be definitely tackled within the aerogel community. Venues for progress and current gaps to be filled in aerogel technology are identified and intense research efforts are needed from different approaches and domainsThere are already incipient international collaborative efforts on research, training,[29,194] and events specifically focused on the cross-fertilization of ideas on this environmental aspect for aerogels, as well as innovation grants funded by public funding bodies[195,196] to boost business plan initiatives with efficient communication between innovators, industrial players, and business developers.

Biographies

Prof. Carlos A. García-González received a PhD in Process Engineering in 2009 from the Technical University of Catalonia (Spain), complementing his education during a two-year postdoctoral stay at the Hamburg University of Technology (Germany) and a two-year industrial experience at the company Solvay (Brussels, Belgium). In 2014, he moved to the Department of

Pharmacology, Pharmacy and Pharmaceutical Technology of the University of Santiago de Compostela (Spain). His research within the AerogelsLab team (aerogelslab.egd.gal) is focused on the development of novel processing approaches and sustainable solutions using green technologies to obtain nanostructured materials validated for biomedical purposes. He places particular emphasis on aerogel-based materials intended for drug delivery, wound healing and regenerative medicine.

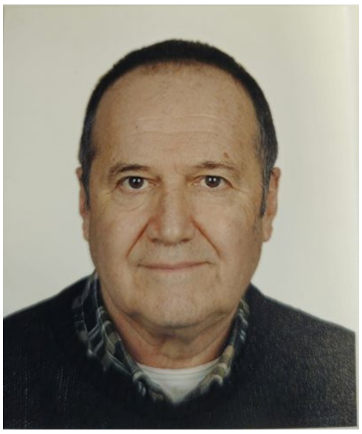
María Blanco-Vales graduated in Chemistry from the University of Santiago de Compostela. She is currently working on her PhD thesis at the same university on technological innovations in biomedical materials towards their efficient and sustainable production. She is particularly focused on the reprocessing of biopolymer-based aerogels and on the production of aerogels using different sources of waste as precursors.

Joana Barros (JB) earned a PhD (Cum Laude) in Biomedical Engineering at the Faculty of Engineering of the University of Porto (FEUP), in collaboration with the Institute for Biomedical Engineering (INEB)/Institute for Research and Innovation in Health (i3S), and Univ. Minho (UM). She is currently working as a Junior Researcher (CEECIND, 5th edition) in i3S's Bioengineering Surfaces Group. Her research has focused on the development of multifunctional approaches based on anti-infective biomaterials and drug delivery systems as biological and bioactive platforms for effective human infection treatment.

Dr A. C. Boccia received her PhD degree in chemistry in 2004 from the University of Salerno. From 2004 to 2005, she worked as a researcher at Basell Polyolefins in Italy and France. Since 2006, she has been conducting research at the CNR-National Research Council in Milan. Her research is focused on the synthesis and microstructural characterization of plastic polymers for different applications, by NMR spectroscopy. Recently, she has broadened her research interests to the development of natural polymers from polysaccharides, particularly for the production of cryogels used in life sciences and packaging. Additionally, Dr. Boccia is contributing to the field of cultural heritage, applying NMR spectroscopy to investigate the degradation mechanisms of plastic artworks and developing strategies for their preservation.

Tatiana is an expert in polymer chemical physics, in particular, in polymer solutions, gels and aerogels, with a focus on biomass-based polymers, and also in polymer composites with natural fibers. She defended her PhD at the Institute of Macromolecular Compounds of Russian Academy of Sciences and got her Habilitation in France. Tatiana holds “research director” position in the Center for Materials Forming of Mines Paris, France. She is also one of the editors-in-chief of Carbohydrate Polymers journal and chair of Cellulose and Renewable Materials Division of American Chemical Society. She published more than 180 articles, and in 2020, she was awarded a “Silver Medal” by CNRS (France) for the outstanding research on bio-aerogels.

Prof. Luisa Durães received her PhD in Chemical Engineering in 2008 from the University of Coimbra, Portugal. Currently, she is an Associate Professor at the same university, in the Department of Chemical Engineering. Her research interests focus on the synthesis, functionalization, reinforcement, and scale-up of nanostructured materials, mainly aerogels of several oxides and polysaccharides, for application in the environmental, energy and biomedical sectors. Recently, she has also been developing upcycling strategies for the production of aerogels and coatings from plastic waste.



Prof. Dr. Can Erkey received his B.S. in Chemical Engineering from Boğaziçi University in 1984 and his Ph.D. in Chemical Engineering from Texas A&M University in 1989. He started his academic career at the Chemical Engineering Department of the University of Connecticut in 1995 as an assistant professor. He then joined the Chemical and Biological Engineering Department at Koç University in 2006. His research interests are in hydrogen technologies, nanostructured materials and supercritical fluids. He has been working in the field of aerogels since 2003 with an emphasis on electrocatalytic and adsorptive applications related to hydrogen fuel cells, electrolyzers and gas separation systems.

Marta Gallo is a biomedical engineer holding a PhD in Material Science. She first worked on ceramics for bone replacement. Lately, she broadened her expertise by getting involved in the development of silica-based porous systems, including aerogels, for the adsorption or the release of molecules in the environmental and pharmaceutical sectors, respectively. Her know-how encompasses the development of these systems from their synthesis up to the evaluation of their performances. Marta Gallo carried out her Ph.D. and worked as a post-doc in France (INSA, Lyon) and in Germany (FAU University, Erlangen); she now works in Italy at Politecnico di Torino.

Dr. Petra Herman obtained her PhD in 2021 at the University of Debrecen and joined the faculty of this university in the same year as an assistant professor. She has been working in the field of environmental chemistry, and her research is mainly focused on the development of biopolymer aerogels for environmental remediation and the recovery of valuable metal compounds.

Dr. József Kalmár earned his PhD in 2013 at the University of Debrecen, where he is now an associate professor. He established a new research direction focusing on exploring application-related structure-properties-function relationships in porous nanostructured materials. His team has described the intimate mechanisms of hydration-induced structural changes in aerogels, sorption equilibria, catalytic reactions, and surface complexation of metal ions. His novel mechanistic approach has been utilized in several joint EU projects.

Ana Iglesias-Mejuto, PhD in Pharmaceutical Technology from the University of Santiago de Compostela, completed her PhD Thesis focused on the 3D-printing of bioaerogels for bone tissue engineering applications and also on their evaluation by different physicochemical and biological tests. She is currently working on the development of bioinks for the 3D-printing of hydrogels for regenerative medicine purposes.

Dr. Wim Malfait completed his PhD in Earth Science from ETH Zurich in 2007 on the molecular structure of silicate glasses and melts. He then worked at Okayama University and ETH Zurich on the deep Earth's interior using synchrotron high-pressure experimentation. In 2013, he moved to Empa and into the aerogel field, where he now heads the Laboratory for Building Energy Materials and Components, working on silica, biopolymer and polyimide aerogels, carbon capture, and bio-based thermal insulation.

Dr. Shanyu Zhao is the group leader of the Functional Aerogel Materials Group at the Building Energy Materials and Components Laboratory at the Swiss Federal Laboratories for Materials Science and Technology (Empa). He received his PhD in Materials Science from Dalian University of Technology (2011), and during his PhD, he worked as a visiting researcher in the Department of Chemistry at Brown University (2009). He has extensive experience in scientific research and product development, with a strong focus on technical project management in the areas of functional porous and energy materials. His research interests include sol–gel chemistry, additive manufacturing, and the thermal, biomedical, and environmental applications of aerogels and nanocomposites.

Lara Manzocco is an Associate Professor of Food Technology at the University of Udine. Her research activity is relevant to the use of innovative non-thermal technologies to steer stability, physical structure, and techno-functional properties of food products. In this context, she developed novel processes and protocols for the preparation of food-grade mesoporous aerogels to be used as delivery systems and structuring agents.

Prof. Stella Plazzotta received her PhD in Food and Human Health in 2019 from the University of Udine (Italy). She is currently working as an Associate Professor at the University of Udine. She has been devoted to the development of sustainable technological strategies for the upcycling of by-products from the food industry into value-added ingredients and materials for the food sector.

Stoja Milovanovic obtained her master’s degree in Biochemical Engineering and Biotechnology (in 2010) and her doctorate in Chemical Engineering (in 2015) from the Faculty of Technology and Metallurgy (University of Belgrade, Serbia) where she currently works as a Senior Research Associate. Her work primarily focuses on supercritical carbon dioxide-assisted processes, such as extraction from plant material, as well as polymer drying, foaming, and impregnation, to develop new or improved products and materials. She places particular emphasis on sustainable and green processing methods tailored for the pharmaceutical, food, and medical sectors.

Prof. Monica Neagu received her PhD degree in 1996 from the Romanian Academy of Science. She is currently the Head of Immunology Laboratory “Victor Babes” National Institute of Pathology in Bucharest, Romania, and a Habilitated Professor of Immunology at the University of Bucharest. In recent years, she has been involved in immunotoxicology studies of various compounds to be used in the biomedical domain.

Dr. Loredana E. Nita, is a 1st degree researcher at the "Petru Poni" Institute of Macromolecular Chemistry since the year 2001. In April 2008 she received her PhD degree in Chemistry. In 2019 she obtained the title of doctoral supervisor (habilitation) and since 2020 she supervises 4 PhD students. Her activity was performed both from a fundamental as well as an applied standpoint, using a multidisciplinary approach, allowing her to develop the following research directions: - obtaining pH and thermo-sensitive hydrogels by adjusting the chemical functionality of the gel structure through the inclusion of a second interpenetrating network and/or specific entrapped structures, - obtaining hydrogels with a multi-membrane organization through a multi-stage gelation process; - obtaining and testing systems that have encapsulated drugs, starting from advanced functional macromolecular structures made by self - assembling process; - testing the possibilities for the use of hydrogels as a controlled drug delivery system.

Prof. Patrina Paraskevopoulou received her PhD in Chemistry in 2003 from the National and Kapodistrian University of Athens (NKUA). From 2006 to 2007, she worked as a postdoctoral fellow at the Missouri University of Science and Technology (MS&T) in the USA. In 2017, she was a Visiting Professor at the Hamburg University of Technology (TUHH), Germany, funded by DAAD through its Research Stays for University Academics and Scientists program. She is currently a Full Professor in the Department of Chemistry at NKUA, Greece. Her primary research interests focus on inorganic and hybrid organic/inorganic nanostructured materials, such as metal-doped synthetic polymers, biopolymeric and carbon aerogels, and their potential applications in energy, environmental remediation, catalysis, and biomedicine.

Prof. Anna Roig holds a degree in Physics and received a PhD in Materials Science from the Universitat Autònoma de Barcelona (UAB), complementing her education at the Royal Institute of Technology in Stockholm and Northeastern University in Boston. Currently, she works at the Materials Science Institute of Barcelona (ICMAB-CSIC), where she leads the Nanoparticles and Nanocomposites Group (nn.icmab.es). Her research pivots around the rational synthesis of nanoparticles and nanocomposites using green chemistry and biotechnology routes while validating those materials for biomedical applications. Specifically, her group has developed nanomaterials as drug delivery vehicles, contrast agents, and bio-based hydrogels/aerogels.

Dr. Rosana Simón-Vázquez received her PhD in 2009 from the Autonomous University of Barcelona, Spain. In 2010, she joined the University of Vigo (Spain) as a postdoctoral researcher. From 2014 to 2016, she undertook a research secondment at the Institut Galien, University of Paris Sud (France), and in 2017, at the Center for Research in Molecular Medicine and Chronic Diseases (Santiago de Compostela, Spain). She is currently working as an associate professor at the University of Vigo. Her research interests are primarily focused on nanotoxicology, nanomedicine, and immunotherapy.

Since 2008 Irina Smirnova is a Professor at the Hamburg University of Technology, Hamburg, Germany. She serves as the Head of the Institute of Thermal Separation Processes and as the Vice-president of research of the TUHH. Her research interests include aerogels, high pressure processes, thermodynamics and separation technologies. She has published over 200 papers in these fields (https://orcid.org/0000-0003-4503-4039). Before that, the engineer was a visiting scientist at Sogang University in South Korea and worked until 2008 as a group leader and postdoctoral candidate at the Institute for Thermodynamics and Thermal Process Engineering at the Technical University of Berlin and at the Institute for Thermal Process Engineering at the University of Erlangen-Nuremberg. Smirnova studied physical chemistry at the State University of St. Petersburg and received her doctorate in 2002 from the Technical University of Berlin on the synthesis and application of aerogels in process engineering. She completed her postdoctoral qualification in 2008 at the University of Erlangen-Nuremberg. In her scientific career, she has also been awarded the DECHEMA Young Academic Award, the Hamburg Teaching Award and the Ralf Dahrendorf Prize.

Željko Tomović obtained his BSc and MSc degrees in Chemistry at the University of Kragujevac, Serbia. In 2001, he joined the group of Prof. Klaus Müllen at the Max Planck Institute for Polymer Research in Mainz, where he completed his PhD thesis in 2005. After a postdoc in the group of Prof. Bert Meijer at the Eindhoven University of Technology, he joined BASF in 2006, working in the field of polyurethane and carbon-rich materials research. In 2020, he became a full professor at the Eindhoven University of Technology. His research interests center on circular and high-performance polymer materials.

Dr. Clara López Iglesias obtained her PhD in Drug Research and Development at the University of Santiago de Compostela in 2020. She then obtained a 3-year postdoctoral fellow Xunta de Galicia, which included 2-year research stay at the Free University of Berlin. Currently, she holds another Xunta de Galicia postdoctoral fellowship and works as a postdoctoral researcher at the University of Santiago de Compostela. Her research interests include aerogels, supercritical fluids, drug delivery, nanomedicine, and biomaterials.

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Non-technical synopsis

Aerogels are lightweight materials well-positioned to support industry with more efficient use of resources as demand for eco-conscious technologies rises.