

Review and Perspectives on the Sustainability of Organic Aerogels

Original

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Review and perspectives on the sustainability of organic aerogels

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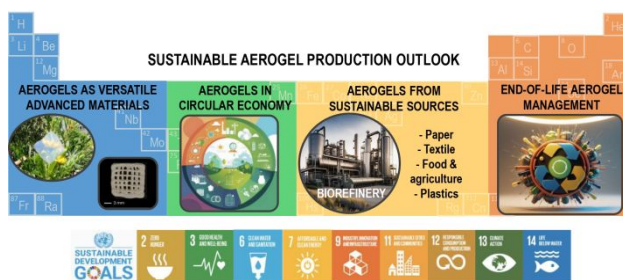
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24 53

25 54 **TOC graphic**



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3 **57 Abstract**
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5 58 Aerogels are exceptionally lightweight materials characterized by their high open porosity and
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7 59 remarkable specific surface area currently used across a wide array of industrial sectors, from
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9 60 construction to energy storage, and have great potential for expanding their applicability and
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11 61 unlocking new market opportunities. Driven by global economic growth and an intensifying
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13 62 environmental crisis, there is a growing demand for engineering innovations that prioritize
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15 63 sustainability. Aerogels are well-positioned to support these sustainability efforts. Their unique
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17 64 properties make them ideal for energy-saving solutions, environmental remediation, and more
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19 65 efficient use of resources. As the demand for eco-conscious technologies rises, aerogels are
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21 66 poised to contribute significantly to the development of greener, more efficient products and
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23 67 processes across multiple industries. The sustainability of aerogel technology is crucial for the
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25 68 mid-to-long-term future, yet its current status has been scarcely reviewed in the literature. This
26
27 69 article explores and critically reviews significant advances on organic and hybrid aerogels in the
28
29 70 current socioeconomic scenario with selected case studies endorsing their contribution to the
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31 71 UN Sustainable Development Goals. It also identifies research gaps while proposing innovative
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33 72 strategies to enhance the sustainability of aerogel production through the application of circular
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35 73 economy principles. Key strategies discussed involve the fabrication of aerogels using eco-
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37 74 friendly sources, such as biopolymers derived from biorefinery processes or from waste
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39 75 materials. Additionally, this article examines the development of methods for the reuse,
40
41 76 recycling and end-of-life management of aerogels, along with the implementation of more
42
43 77 efficient processing routes. Ultimately, this work highlights the need for comprehensive
44
45 78 assessments of aerogel sustainability by through life cycle assessment (LCA), and evaluations of
46
47 79 safety and toxicity. By addressing these critical aspects, the potential of aerogels to contribute
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49 80 to a more sustainable future appears highly favorable from both commercial and research
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51 81 perspectives, paving the way for a circular aerogel economy and providing a lasting impact to
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53 82 the society we live in.
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84 **Keywords:** bio-aerogels; circular technologies; sustainable production; aerogel recycling; waste

85 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

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3 112 These new regulations, as well as market and societal demands, require the development of
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5 113 sustainable, innovative and advanced functional materials. A prominent class of materials that
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7 114 could address some of the mentioned challenges are aerogels, nanostructured materials
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9 115 endowed with unique properties e.g., high specific surface area (usually above 100 m²/g), low
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11 116 bulk density (usually below 0.2 g/cm³) and open porosity (usually higher than 85% with high
12
13 117 presence of interconnected mesopores). Aerogels are especially attractive for a wide range of
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15 118 applications, from thermal insulation in buildings and industrial facilities,¹⁴ to environmental
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17 119 (adsorbents for air, soils,¹⁵ and water remediation,^{16,17} selective binders for CRMs recovery,^{18,19}
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19 120 sensors and catalysts,^{20,21} sound absorbers²²) and biomedical uses (scaffolds for regenerative
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21 121 medicine,²³ thermal insulators for photothermal oncotherapy,²⁴ dressings for wound healing,²⁵
22
23 122 drug carriers²⁶). In addition, aerogels can be found in emerging applications in the food sector,
24
25 123 where they may act as packaging materials with advanced functionalities (e.g., cushioning effect,
26
27 124 thermal insulation, release or absorption of desired/undesired compounds) or food ingredients
28
29 125 (e.g., fat replacers, delivery systems for active compounds, etc.).^{27–29} The superior properties of
30
31 126 aerogels in different fields resulted in a high scientific impact, such that the prestigious authority
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33 127 IUPAC (International Union of Pure and Applied Chemistry) identified aerogels as one of the Top
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35 128 Ten Emerging Technologies in Chemistry in December 2022.³⁰
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42 129 From an industrial point of view, the aerogel market is estimated at 1,155 million USD by 2025,
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44 130 with an average annual growth rate of 26% until 2030.³¹ Advances in recent years encompass
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46 131 the use of various sources (inorganic materials, organic synthetic polymers, natural polymers,
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48 132 carbons, hybrid materials) for single component or composite aerogels, diverse morphologies
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50 133 (powder, beads, monoliths, mats, boards, films) and dimensions (from nanometers to meters),
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52 134 along with modelling, production scale-up, and health and safety assessments.
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56 135 From a sustainability point of view, aerogel producers should decide on an energy-efficient
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58 136 drying process, probably the most critical step of the production line, with consideration on the
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3 137 material source and the intended use and performance. Furthermore, rational use of resources
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5 138 should include minimization of materials use, reuse of solvents, and recycling of unspent
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7 139 precursors, towards zero-waste in the production line. These must be also contemplated for the
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9 140 pretreatment of raw materials (e.g., extraction, derivatization, milling, purification) and the
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11 141 post-processing of aerogels (e.g., polishing, cutting, milling, carbonization, sterilization).
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13 142 Production costs can be optimized through the reduction of cycle time (for batch production),
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15 143 the increase of throughput (for continuous production), and the integration of processes.
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17 144 Studies on most of these technological aspects have been recently reviewed in the literature,^{32,33}
18
19 145 but life cycle and sustainability assessment studies are still scarce.^{34,35}
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23 146 While minimizing aerogel manufacturing time and costs, business strategies should also focus
24
25 147 on sustaining and further extending the niche markets for this material. This perspective also
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27 148 addresses the circularity of different wastes that may feed the production of aerogels with an
28
29 149 increased circular material use rate, reduced usage of raw materials, and lower energy
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31 150 consumption. For instance, developing more sustainable thermal insulation products could have
32
33 151 important environmental and economic impacts as the building thermal insulation market is
34
35 152 projected to almost double in the next ten years (from USD 25 billion in 2022 to USD 45 billion
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37 153 in 2032).³⁶ Similarly, the introduction of circular economy principles by using waste products in
38
39 154 the wound healing domain will have an enormous economic impact, as the global wound care
40
41 155 market is expected to expand to 28 billion USD by 2029. Similarly, aerogels intended as food
42
43 156 grade oil structuring ingredients will position in the market of fat replacers, which reached 2.2
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45 157 billion euros in 2022 in Europe and with an annual growth rate of *ca.* 6%.
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51 158 The societal impact will be tied to both the lowering of the country's consumption footprint and
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53 159 its raw material self-sufficiency. However, sustainability and end-of-life management of
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55 160 aerogels, as well as rational use of resources for their production, have received little attention
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58 161 thus far, providing a technological and commercial challenge to the current state-of-the-art.
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3 162 Sustainable aerogel production has recently evolved, with significant and exponential research
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5 163 growth rates.³⁷ The expected roadmap for this topic from different perspectives and approaches
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7 164 will be discussed in the following sections of this article: (i) identifying aerogels positioning in
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9 165 the United Nations (UN) Sustainability Development Goals (SDG) context (Section 2); (ii)
10 166 performing a critical review of significant advances and searching for research gaps in innovative
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12 167 and (still) underexplored use of sustainable sources for aerogel production from biorefinery
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14 168 (Section 3), (iii) addressing wastes and by-products (Section 4); (iv) proposing various options for
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16 169 end-of-life of aerogel wastes by their recycling, reprocessing or upcycling (Section 5); and (v)
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18 170 proposing the implementation of process integration strategies and emerging technologies in
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20 171 aerogel production to minimize the consumption of resources and decrease energy use (Section
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22 172 6). Finally, end remarks and other remaining challenges in the sustainable production of aerogels
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24 173 are compiled and emphasized (Section 7).

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175 **2. Aerogels in the UN SDGs context**

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

183 Bio-based aerogels (or bio-aerogels) obtained from natural polymers are advantageous for their
184 abundance, biocompatibility, and biodegradability.^{28,29} Bio-aerogels are in harmony with the
185 SDGs due to their inherent degradability, which ensures their disposal in accordance with the
186 circular economy. Biocompatibility is another intrinsic feature of bio-aerogels that opens the
187 sustainable application of these solutions in the biomedical, environmental, food, and packaging

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3 188 fields, aligning with SDG3 (good health and well-being) (Fig. 1). From one side, these eco-friendly
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5 189 solutions are sustainable alternatives to standard non-renewable medical or packaging
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7 190 materials, which are reducing environmental impact directly throughout their life cycle. On the
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10 191 other hand, biocompatibility makes bio-based aerogels food-grade materials, supporting their
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12 192 use as ingredients for sustainable and healthier diets. These sustainable applications of aerogels
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14 193 are being studied using different renewable sources such as chitosan, cellulose, poly(lactic acid),
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16 194 proteins, etc.^{39,40} These derivatives could be obtained by the valorization of agricultural wastes
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18 195 (e.g., crustacean shell or sugarcane) or food industry by-products,⁴¹ thus fulfilling the SDG12
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20 196 (responsible consumption and production) and the SDG2 (food security, improved nutrition and
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22 197 promotion of sustainable agriculture). Additionally, aerogel sorbents manufactured by the
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24 198 valorization of agricultural and food wastes, or aerogels obtained from mineral phases (e.g.,
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26 199 silica and clay), are sustainable alternatives with high capacity for water remediation and
27
28 200 recovery of CRMs, such as Pt, Pd, Ag, Au, lanthanides or actinides from aqueous media.^{42–44} In
29
30 201 this regard, aerogels may contribute to SDG6 (clean water and sanitation) as well as to SDG14
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32 202 (life below water).

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36 203 Buildings and their facilities must have minimum energy requirements since the publication of
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38 204 the *2010/31/EU Energy Performance Building Directive* by the EU. Aerogels represent a new
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40 205 generation of thermal insulation materials with remarkable engineering applications due to
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42 206 their lightweight and extremely low thermal conductivity, as their nanostructure restricts the
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44 207 conductive heat transfer and the movement of gas molecules. Research and development on
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46 208 aerogels as new superinsulating materials is a way to commit to this policy with the ultimate
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48 209 goal of designing net-zero energy buildings. All these efforts are well linked with the SDG11
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50 210 (sustainable cities and communities) (Fig. 1).⁴⁵ As an example, silica aerogels are extremely
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52 211 lightweight and highly effective thermal insulators with a thermal conductivity significantly
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54 212 lower than other commercial insulating solutions. This advantageous skeleton has fostered their
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56 213 use in green building construction or clothing, energy production, automotive, aerospace and
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3 214 military industries thus being in line with the SDG7 (affordable and clean energy) (Fig. 1).^{46,47}
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5 215 Other applications of silica aerogels are being explored like their use as acoustic absorbers due
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7 216 to the low velocity of the sound transmission throughout the matrix of the aerogel thereby
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10 217 promoting the SDG9 (industry, innovation and infrastructure).⁴⁸ In these applications, research
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12 218 should focus on improving the mechanical, processability, and stability properties through the
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14 219 use of organic (polyurethane⁴⁹ or polyimide⁵⁰), carbon⁵¹ or hybrid (silica-organic⁵²) aerogels, as
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16 220 they remain as the biggest challenges for aerogels efficiency and long-term performance.^{53,54}
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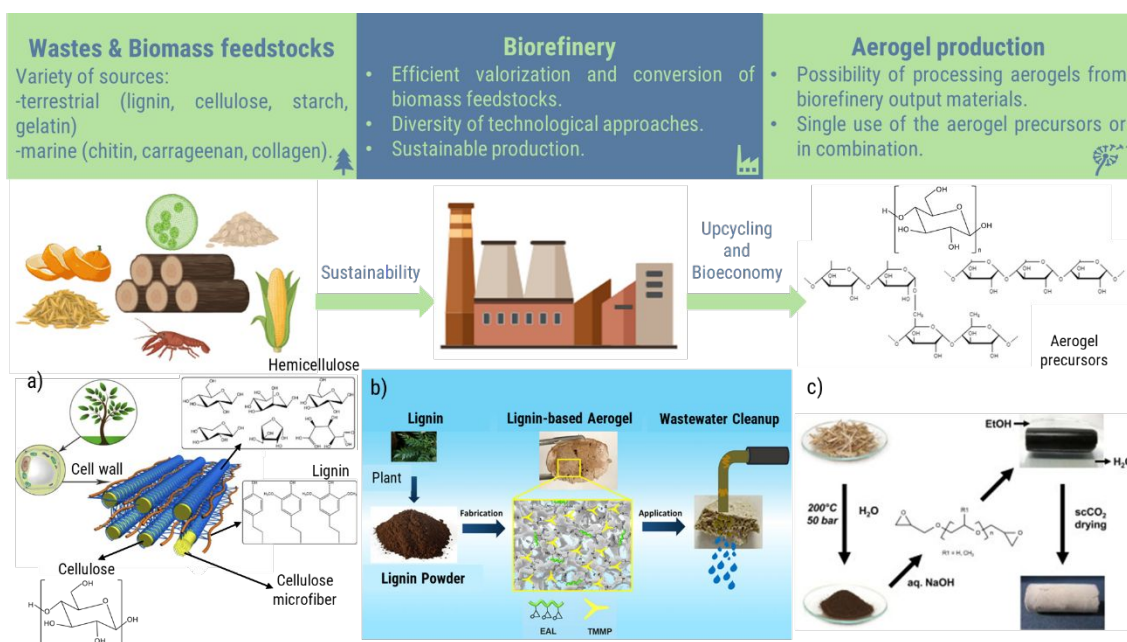


222
223 Figure 1. Outlook of the most remarkable contributions of aerogels to the UN sustainable
224 development goals.

225 226 3. Aerogels from a biorefinery approach

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

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237 Figure 2. Implementation of biorefinery approaches in aerogel production: (a) Lignocellulosic
 238 biomass chemical composition.⁵⁵ Adapted with permission from ref.⁵⁹. Copyright ©
 239 2017, Elsevier; (b) Lignin-based aerogel for wastewater remediation.⁶⁰ Adapted with permission
 240 from ref.⁶⁰. Copyright ©2022, Elsevier; (c) Aerogel preparation from lignin derived from wheat
 241 straw.⁶¹ Reprinted with permission from ref.⁶¹. Copyright ©2014, Elsevier.

242 Biorefineries employ several technological approaches, the most important of which depend on
 243 biomass feedstock attributes (e.g., origin and amount of residues) and product standards targets
 244 (yield, purity). In the bio-based economy paradigm, using these biorefinery output materials to
 245 make aerogels can contribute to upcycling into high added-value advanced materials. The
 246 sustainable production of materials from biomass should be supported not only by the source
 247 itself but also by using eco-friendly and safe production technologies with a low CO₂ footprint,
 248 avoiding the use of hazardous reagents, and preventing mass losses during the process cycle.
 249 Aerogel end-of-life and waste management should also be considered (cf. Section 5), as they
 250 should be safe for producers and customers, sustainable for the ecosphere, and economically
 251 feasible. The management of aerogel leftovers after use should be defined by design; otherwise,
 252 the sustainable production approach will be diluted or neglected.

253 Two main biorefinery sources can be used to produce aerogels (bio-aerogels): terrestrial and

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3 254 marine tissue wastes. The most common raw materials from terrestrial vegetal wastes are lignin,
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5 255 cellulose, pectin, and starch, while silk fibroin, gelatine, whey protein and keratin are from
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7 256 terrestrial animal wastes and by-products.^{62–69} Agarose, chitin and derivatives (chitosan),
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10 257 carrageenan, and collagen and derivatives (gelatine) are the most used raw materials from
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12 258 marine wastes.^{62,67,70,71} These raw materials are used alone or in combination with natural or
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14 259 synthetic components, for example, crosslinkers or other admixtures (e.g., nanoparticles), to
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16 260 tune the physicochemical properties of aerogels. It should be noted that the use of biorefinery-
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18 261 derived aerogels may be limited in some areas, such as in life science applications (biomedicine,
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20 262 pharmaceuticals), due to the type, quality, and variability of the biorefinery source.^{26,27} Finally, the
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22 263 choice of additives for the material design should be rational to avoid the underscoring of
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24 264 aerogel sustainable production.
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28 265 29 266 **4. Aerogels as a green way to waste upcycling**

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31 267 Economic development, population growth, and fast urbanization are associated with a
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33 268 significant increase in consumption and a consequent increase in waste. Currently, 2.1–2.3
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35 269 billion tons of only municipal solid wastes per year are generated worldwide⁷² and the global
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37 270 circular material use rate is generally low (e.g., only 11.5% in 2022 in EU-27)⁷³. This translates
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39 271 not only in high managing and disposal costs but also in wasting of resources (land, water, and
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41 272 energy) necessary to produce goods.
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45 273 Waste recycling refers to the reuse of existing waste material, which frequently results in lower-
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47 274 quality products with limited applications. The concept of waste upcycling refers to its reuse to
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49 275 fabricate upgraded or added-value materials, also known as waste valorization.⁶⁴ Aside from the
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51 276 transformation of the waste into relatively pure raw materials via biorefinery processes (cf.
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53 277 Section 3), waste upcycling can also be accomplished by simpler processing involving the integral
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55 278 conversion of the pure biomass into valuable derivatives.^{74,75} Because no strong purification or
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57 279 extraction is performed in this situation, the high value of the resulting materials can only be
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3 280 achieved through complex supra- or macromolecular structures or architectures.^{76,77} As
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5 281 previously mentioned, aerogels are regarded as high-value materials with outstanding textural
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7 282 properties. However, applications such as food and beverage (direct) packaging, cosmetics, or
8
9 283 technical clothing are usually not considered for waste-derived aerogels,⁷⁸ due to a general
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11 284 lower purity than those obtained through biorefinery.

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15 285 A first important step to allow for the usage of integral biomass as aerogel precursor would be
16
17 286 waste upgrade from waste to by-product. According to the waste framework Directive (Directive
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19 287 2008/98/CE), a waste ceases to be a waste and is classified as a by-product if specific conditions
20
21 288 are met. For instance, the conversion of wasted biomass into food-grade aerogels would
22
23 289 necessitate the establishment of a dedicated production chain, commencing with waste
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25 290 collection. The latter should adhere to rigorous food regulations to ensure the minimization of
26
27 291 safety risks. In this context, it should also be pointed out that in most cases, biomass waste
28
29 292 produced by food industries or consumers is currently classified as a by-product since it is
30
31 293 utilized in biogas production, composting or animal feeding.⁷⁹ However, these strategies result
32
33 294 in products of diminished value in comparison to those reaching the market. By contrast, the
34
35 295 use of food by-products as aerogel precursors would allow their upcycling leading to high value
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37 296 materials.

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42 297 Nguyen and coworkers conducted a thorough analysis of aerogels produced from waste
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44 298 materials,³⁸ focusing on the main outcomes of the research, such as aerogel characterization
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46 299 and performance. Conversely, in this section, an overview of the process will be provided, from
47
48 300 the collection and processing method of the pure biomass into aerogels to their final
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50 301 performance and applicability of the resulting aerogels. The environmental impact of the whole
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52 302 process, from waste production to final reuse or disposal, will be discussed and research gaps
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54 303 and future directions will be identified.

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58 304 **4.1. Waste-based or byproduct-based aerogel preparation**
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3 305 Aerogels can be produced from waste or by-products that are dissolved or dispersed in a suitable
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5 306 medium, after chemical or physical treatment that will promote the formation of a 3D network.
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7 307 The mixture of textile fibers waste with a standard silica sol is an option to obtain composite
8
9 308 aerogels with improved thermal and acoustic insulation properties, high water-contact angle
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11 309 and high flexibility.⁸⁰ Another example is the dissolution of recycled tire granulate rubber by and
12
13 310 oxidant acid to form a rubber sol that can be easily mixed with the silica sol before its gelation.
14
15 311 In this way, a hydrophobic efficient thermal insulator can be prepared, formed by a continuous
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17 312 rubber-silica aerogel matrix.⁸¹ It can also be extended to compound other inorganic or organic
18
19 313 sol-gel-derived phases. In some cases, phase separation and gelation can be induced by a non-
20
21 314 solvent, as for example for aerogels prepared from cellulose-based textile.^{82,83} Alternatively,
22
23 315 waste-derived aerogels can be produced by direct mixing waste or by-product materials with
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25 316 EtOH or water/EtOH mixtures with increasing concentrations of EtOH⁸⁴ followed by supercritical
26
27 317 drying. In this way, water originally present in the waste material is substituted with ethanol,
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29 318 which is then removed via supercritical drying. This process is particularly convenient when the
30
31 319 waste material already presents a natural architecture (e.g., plant wastes and by-products)
32
33 320 suitable for aerogel production.

321 **4.2. Waste streams**

322 *Food and agriculture waste*

323 According to the FAO 2024 “Global Facts” (FAO, 2024),⁸⁵ around 8-10% of global greenhouse gas
324 emissions are associated with food that is not consumed with *ca.* 931 million tons lost in the
325 supply chain, from after harvest, and prior to reaching retail shelves in 2021, and 1.05 billion
326 tons of food wasted in households, food services and retail in 2022. This represents *ca.* 30% of
327 all food produced worldwide for human consumption raising significant ethical concerns.
328 Food by-products are often represented by animal or plant tissues which are discarded during
329 food processing such as swarfs, substandard materials or exhausted matrices e.g., from fruit
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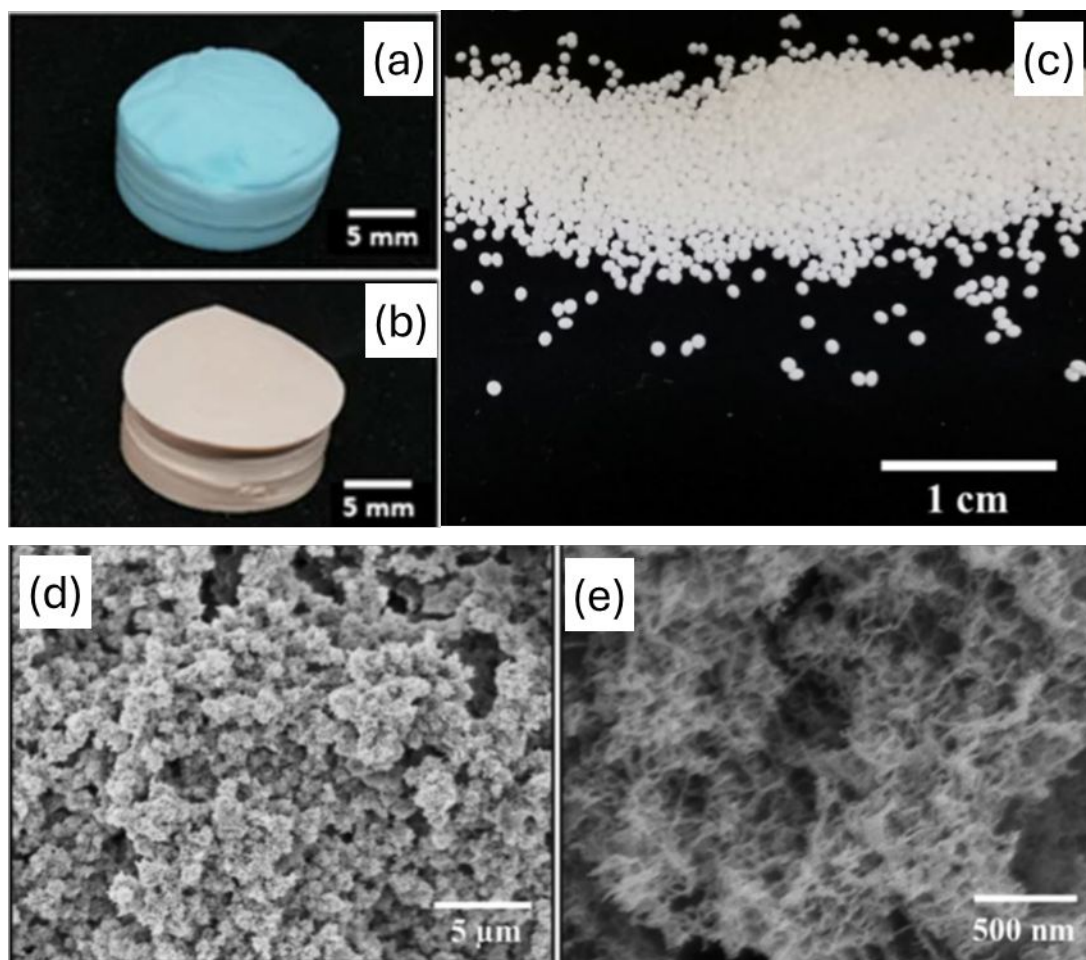
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3 330 juice and oil extraction. These cellular tissues can be treated as gel-like materials made by a
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5 331 complex biopolymer network, mainly structured by cell wall cellulose fibers, embedding water
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7 332 within intra- and inter-cellular spaces. The direct conversion of these “gel-like” materials into
8
9 333 “aerogel-like” ones could thus represent a possible approach to sustainably turn critical biomass
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11 334 into shelf-stable ingredients without further generation of waste. For instance, by-products
12
13 335 (external leaves and core) from industrial fresh-cut processing of iceberg salad were submitted
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15 336 to water-to-ethanol substitution and supercritical CO₂ drying, producing a white aerogel-like
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17 337 material which could be used as packaging, absorbent, or innovative carrier for both lipophilic
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19 338 and hydrophilic compounds.⁸⁴ When the same procedure was applied to homogenized
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21 339 substandard peas, colorless powders without vegetable sensory notes, but with high nutritional
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23 340 value and technological functionality were obtained.⁸⁶
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27 341 Food by-products have also been used to produce fully biodegradable pure⁸⁷ and hybrid⁸⁸
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29 342 aerogels with high porosity. In the latter case, the food residue was homogenized and structured
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31 343 into a superimposed architecture by means of additional gelling agents (e.g., k-carrageenan,
32
33 344 polyvinyl alcohol).⁸⁸ This approach allowed turning discarded salad leaves into aerogels for food
34
35 345 applications. An important limitation of waste-derived natural polymers resides in their high
36
37 346 hydrophilicity, which has derived into strategies for hydrophobic enhancement of the resulting
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39 347 aerogels.⁸⁹ Lignocellulosic aerogels were produced from spent ground coffee and apple
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41 348 pomace⁹⁰ with enhanced hydrophobicity through silanization in a liquid phase or by vapor
42
43 349 deposition. Silanes are common hydrophobizing agents for cellulose, forming polysiloxane
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45 350 structures by reacting with the hydroxyl groups of the cellulose fibers through a condensation
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47 351 reaction.⁹¹ However, it should be noted that Si-O-C bonds are easily hydrolysable in the presence
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49 352 of water.
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55 353 *Textile waste*

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57 354 Besides recent awareness about the use of fast fashion,⁹² reality shows an increased
58
59 355 consumption of textile products from 78 to 103 million tons in the last decade, and this tendency
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3 356 is still growing. Each European citizen discards an average of *ca.* 11 kg of textiles annually, most
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5 357 of which are disposed of in landfills or incinerated.⁹³ Landfill disposal has been forbidden in the
6
7 358 European Union since 2016; incineration leads to the release of harmful chemicals and produces
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10 359 significant CO₂ emissions. The textile recycling process possesses a poor economic value, so
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12 360 research groups have explored engineering solutions to provide added value, such as using the
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14 361 fibers for mechanical reinforcement of aerogels.⁸⁰ In the case of cotton fibers, they can also
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16 362 provide a buffer effect for humidity regulation.

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19 363 About 30% of textile are based on cellulose (cotton, viscose, Tencel). Till now, the main option
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21 364 of upcycling cellulose-based waste textile is dissolving the fabric and spinning fibers or casting
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23 365 films; this work is mainly performed on a laboratory scale or by small companies. Recently it was
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25 366 demonstrated that it is possible to make aerogels from cellulose-based textile waste.^{82,83,94}
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28 367 Cellulose fabric, cotton or viscose was dissolved in ionic liquids, coagulated in water or in
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30 368 ethanol, and dried with supercritical CO₂. The bulk density was from 0.07 to 0.2 g/cm³ and
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32 369 specific surface area from 300 to 400 m²/g. These aerogels could be produced in the shape of
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35 370 monoliths or particles (Figure 3).
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372 Figure 3. Aerogels prepared from waste textile ((a) rayon, (b,c) viscose), in the shape of
373 monoliths⁸² (bulk density 0.1 g/cm³; specific surface area 330 m²/g; porosity > 90%) and beads⁸³
374 (roundness 0.97 – 0.98, density 0.08 g/cm³; specific surface area 400 m²/g; porosity 97%); (d,e)
375 their internal morphology, at different scales and similar for all aerogels, is imaged by scanning
376 electron microscopy.^{82,83} Adapted with permission from ref.⁸³. Copyright © 2024, Springer
377 Nature.

378

379 *Paper waste*

380 Paper is one of the most recycled municipal waste streams and *ca.* 72% of paper and pulp are
381 produced from recycled sources.⁹⁵ However, the recycling process after several cycles causes
382 fiber damage and reduced cellulose molecular weight.⁹⁶ The life cycle of this low-quality paper
383 waste could be extended by its transformation into new and valuable aerogel-based products.
384 Paper waste was used as a carbon source for the fabrication of carbon-based aerogels with
385 outstanding performances as absorbents from water, with sorption rate values at least two

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3 386 orders of magnitude higher than those of activated carbon.⁹⁷ A typical preparation procedure
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5 387 consists in dissolving paper waste in water under stirring to obtain the pulp, drying of the fibers,
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7 388 and thermal treatment under inert atmosphere (including pyrolysis and chemical vapor
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9 389 deposition). These materials achieved exceptional surface area values (up to *ca.* 900 m²/g)
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11 390 finding applications in the recovery of organic pollutants,⁹⁷ antibiotics, and oils/organic solvents
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14 391 from water⁹⁸.

17 392 *Plastic waste*

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20 393 Around 400 million tons of plastic are produced annually worldwide.⁹⁹ Most plastic waste
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22 394 management strategies include direct disposal in landfills/the sea, and combustion, both of
23
24 395 which have an important negative environmental impact. Polyethylene terephthalate (PET) is
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26 396 one of the most common plastic and is used in all sorts of consumer products due to its high
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28 397 stability and resistance to degradation.¹⁰⁰ This results in great interest in developing
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30 398 biodegradable alternatives that are stable enough for packaging applications. Besides the
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32 399 development of sustainable alternative materials, recycling of existing PET plastics is a major
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34 400 concern due to poor economic value and lack of environmentally friendly PET process. Recycled
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36 401 PET fibers can be potentially used to produce aerogels of high-value engineering applications
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38 402 (e.g., thermal insulation, CO₂ capture, and oil spill cleaning), but were only tested as PET cryogels
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41 403 so far.^{100,101}

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47 405 **5. End-of-life of aerogel waste management by their recycling, reprocessing or upcycling**

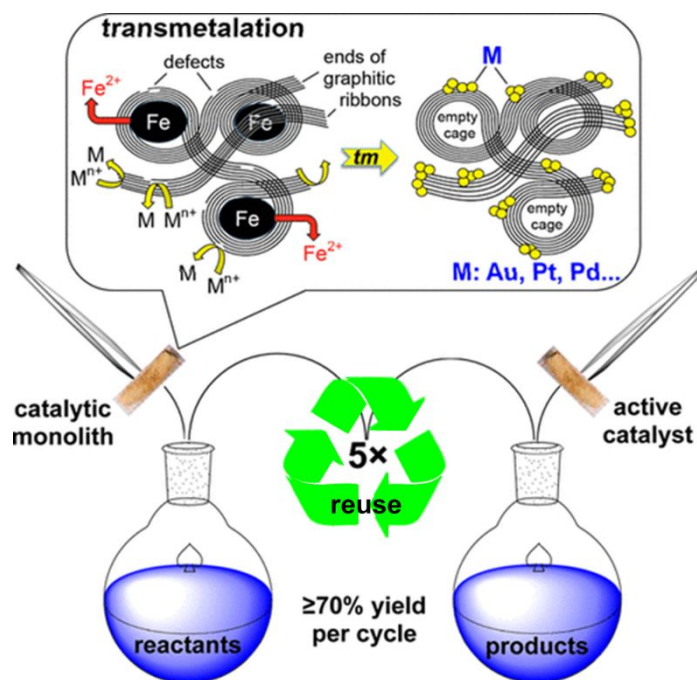
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50 406 Reuse, reprocessing, repurposing and recycling are related concepts in the management of used
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52 407 materials, but with distinct differences. Reuse involves using an item again for the same or a
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54 408 similar purpose without significant modification. Reprocessing involves treating or processing
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56 409 materials to make them suitable for a new use. This often includes physical or chemical changes
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58 410 to return the material to a usable state. Repurposing is the act of using a product or material for
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3 411 a different purpose than it was originally intended, often with little or no alteration. Recycling
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5 412 involves converting waste materials into new materials or products, with the same application
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7 413 or not. All these concepts, especially the first three ones, are in many cases used
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9 414 indistinguishably, as very often reprocessing (also known as regeneration) is a prerequisite for
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11 415 reuse, which can also be considered as a similar, but more general, term to repurposing;
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13 416 recycling also requires reprocessing and/or implies reuse. The application of these concepts to
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15 417 aerogels up to now has been mainly devoted to recycling of the organic solvents used in the
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17 418 synthetic procedures or towards recycling of CO₂ used for supercritical drying of gels. However,
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19 419 recycling, reuse, reprocessing or repurposing of aerogels themselves has not been explored
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21 420 much yet.

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26 421 The reuse or reprocessing of aerogels is an emerging research field for catalysis (i.e., reuse of
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28 422 aerogel catalysts) and environmental remediation (e.g., reuse of aerogel sorbents). It should be
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30 423 pointed out that when it comes to materials used as catalysts or sorbents, sustainability can be
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32 424 assessed based on several factors, including efficiency, longevity, and potential for reuse or
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34 425 recycling. In the context of a circular economy, sorbent materials that can be reused without
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36 426 significant loss of capacity are more sustainable because they can be used over multiple cycles,
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38 427 reducing the need for continuous production and disposal of new adsorbent materials. Although
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40 428 there are only a few such examples compared to the total number of aerogels explored for these
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42 429 specific applications, this can be attributed to the novelty of the concept rather than to the
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44 430 materials being unsuitable for reuse. The number of publications reporting reprocessing/reuse
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46 431 studies has recently increased significantly. This trend will reveal even more aerogel materials
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48 432 that can be reprocessed/reused.

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53 433 In the field of catalysis, monolithic metal-doped carbon aerogels provide an example of easily
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55 434 reusable catalysts.¹⁰² Carbon aerogels bearing metal nanoparticles dispersed homogeneously
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57 435 throughout their volume (Figure 4, M@C aerogels; M: Fe, Au, Pt, Pd, Ni, and Rh), prepared *via*
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59 436 pyrolysis of ferrocene-bearing polyamide aerogels and subsequent transmetallation, exhibited

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3 437 catalytic activity towards (a) oxidation of benzyl alcohol to benzaldehyde (Au@C or Pt@C), (b)
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5 438 reduction of nitrobenzene by hydrazine to aniline (Fe@C) and Heck coupling reactions of
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7 439 iodobenzene with styrene or butyl acrylate (Pd@C), with yields in the range of 85-98%. Due to
8
9 440 their monolithic shape, all these catalysts could easily be removed from the reaction mixture.
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11 441 More importantly, these catalysts were reused five times just by transferring them into a new
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13 442 reaction mixture, without any need for reprocessing. The yields at the end of the fifth cycle
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15 443 in the 70-86% range. Similarly, monolithic Cu@C aerogels, prepared *via* pyrolysis of Cu(II)-chitin
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17 444 aerogels, have been proven to be efficient chemoselective catalysts for the selective reduction
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19 445 of maleimides to succinimides under mild conditions, using hydrosilane as a hydrogen source.¹⁰³
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21 446 Cu@C catalysts could be removed from the reaction mixture *via* filtration and then reused for
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23 447 at least six times without significant loss of activity.
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449 Figure 4. Transmetalation of Fe@C aerogels to M@C aerogels (M: Au, Pt, Pd, Ni, and Rh) leading
450 to reusable catalysts. Adapted with permission from ref.¹⁰². Copyright © 2016, American
451 Chemical Society. (The figure has been edited from its original version, where the authors had
452 used the term "recycle"; in the context of the current understanding of the terminology,
453 "recycle" is replaced with "reuse").

454 In the field of environmental remediation, more examples of reusable aerogels can be found.

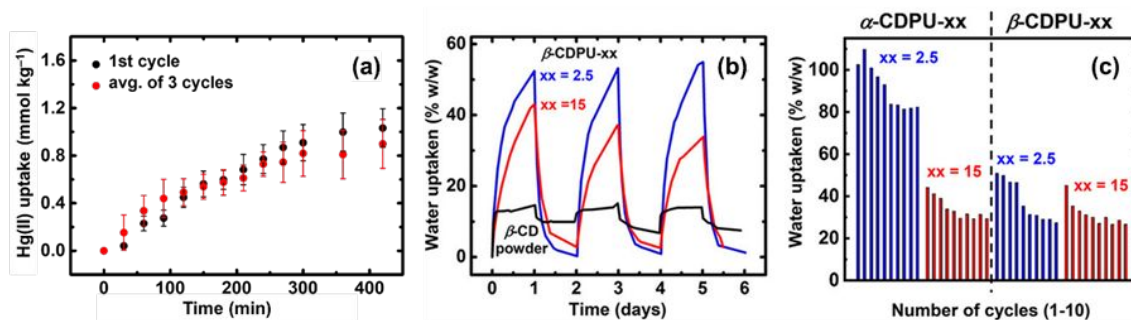
455 For these uses, reprocessing is also needed as the materials after adsorption/absorption of

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3 456 pollutants need to be stripped from the pollutant and regenerated before they can be used
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5 457 again. As demonstrated by the representative examples presented below, the regeneration
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7 458 process can be simple or quite tedious, depending on the materials and the specific application.
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10 459 Crystalline imine covalent organic framework (COF) aerogels, prepared from the reaction of
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12 460 multifunctional amines with multifunctional aldehydes, were tested towards various
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14 461 environmental applications.¹⁰⁴ Depending on their chemical structure and pore size, COF
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16 462 aerogels could be used for the efficient removal of a range of organic solvents (both miscible
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18 463 and immiscible with water), organic dyes and inorganic micropollutants (gold nanoparticles)
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20 464 from water, and for the capture and retention of iodine vapor. COF aerogels with the best
21
22 465 sorption capacities (16-35 times their own weight) for organic solvents, were solvent-exchanged
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24 466 with ethanol and dried again using supercritical CO₂. The reprocessed aerogels exhibited
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26 467 practically no loss of crystallinity or sorption capacity for 10 cycles. Similarly, the ones that
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28 468 showed the best removal efficiency (97%) for methylene blue were washed with acetone and
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30 469 methanol, and they were reused for four more cycles with practically the same performance in
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32 470 the removal of the dye.
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37 471 Biocompatible biopolymer-based aerogels are by design suitable for environmental
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39 472 applications, as they have various potential coordination sites (e.g., -COO⁻, -OH, -NH, -NH₂).
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41 473 One such example is polyurea-crosslinked alginate (X-alginate) aerogels, a new class of materials
42
43 474 recently introduced,¹⁰⁵⁻¹⁰⁷ which can be prepared from pre-formed alginate gels *via* reaction of
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45 475 the functional groups on the surface of the skeletal framework of the alginate (i.e., -OH) with
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47 476 multifunctional isocyanates, leading to the formation of a nano-thin polyurea coating over the
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49 477 entire alginate framework, which enhances the mechanical strength of the materials. Use of
50
51 478 different multifunctional isocyanates allows for tuning of the material properties from the
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53 479 chemical composition perspective.¹⁰⁸ More specifically, X-alginate aerogels derived from the
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55 480 tris(4-isocyanatophenyl)methane (TIPM) are extremely stable in diverse aqueous environments
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57 481 (no swelling, shrinkage, or disintegration has been observed), including seawater. These
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3 482 materials can efficiently uptake organic pollutants (solvents,¹⁰⁹ organic dyes¹¹⁰) and inorganic
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5 483 pollutants, such as U(VI),^{19,111} Pb(II),¹⁰⁹ Eu(III),¹¹² Th(IV),¹¹² Am(III)¹¹¹ and Hg(II)¹¹⁰. Their
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7 484 properties, combined with their high sorption capacity, allow for the reuse of these materials
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9 485 for several cycles. For example, in the case of Pb(II)¹⁰⁹ and Hg(II),¹¹⁰ the materials can be treated
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11 486 with an aqueous solution of Na₂EDTA, washed with water and reused for at least three times
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13 487 (Figure 5a) without significant loss of performance. In the case of U(VI), which is the most
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15 488 extreme case, as X-alginate aerogels adsorb twice their mass (2 g g⁻¹), the materials can be
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17 489 reused for at least five times.¹⁹ Similarly, silica–gelatine hybrid aerogels, prepared via co-gelation
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19 490 of gelatine and tetramethoxysilane, have shown high selectivity for the adsorption of aqueous
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21 491 Hg(II) in the presence of multiple competing ions, e.g., Cu(II), Cd(II), Co(II), Pb(II), Ni(II), Ag(I), and
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23 492 Zn(II) and can be reused for five times, after treatment with Na₂EDTA.¹¹³



493
494 Figure 5. (a) Reusability of X-alginate aerogels for adsorption of Hg(II) ($C_{\text{initial}} = 100$ ppb), after
495 reprocessing that includes washing with aqueous solution of Na₂EDTA and water; (b) Three
496 consecutive cycles of water vapor uptake between a high (99%) and a low (10%) relative
497 humidity environment by β-CDPU-aerogels. (c) Ten consecutive cycles of water vapor uptake
498 monitored every 24 h for α-CDPU- and β-CDPU-aerogels (xx: %w/w concentration of monomers
499 in the sol). (b) and (c) Adapted with permission from ref.¹¹⁴. Copyright © 2019, American
500 Chemical Society.

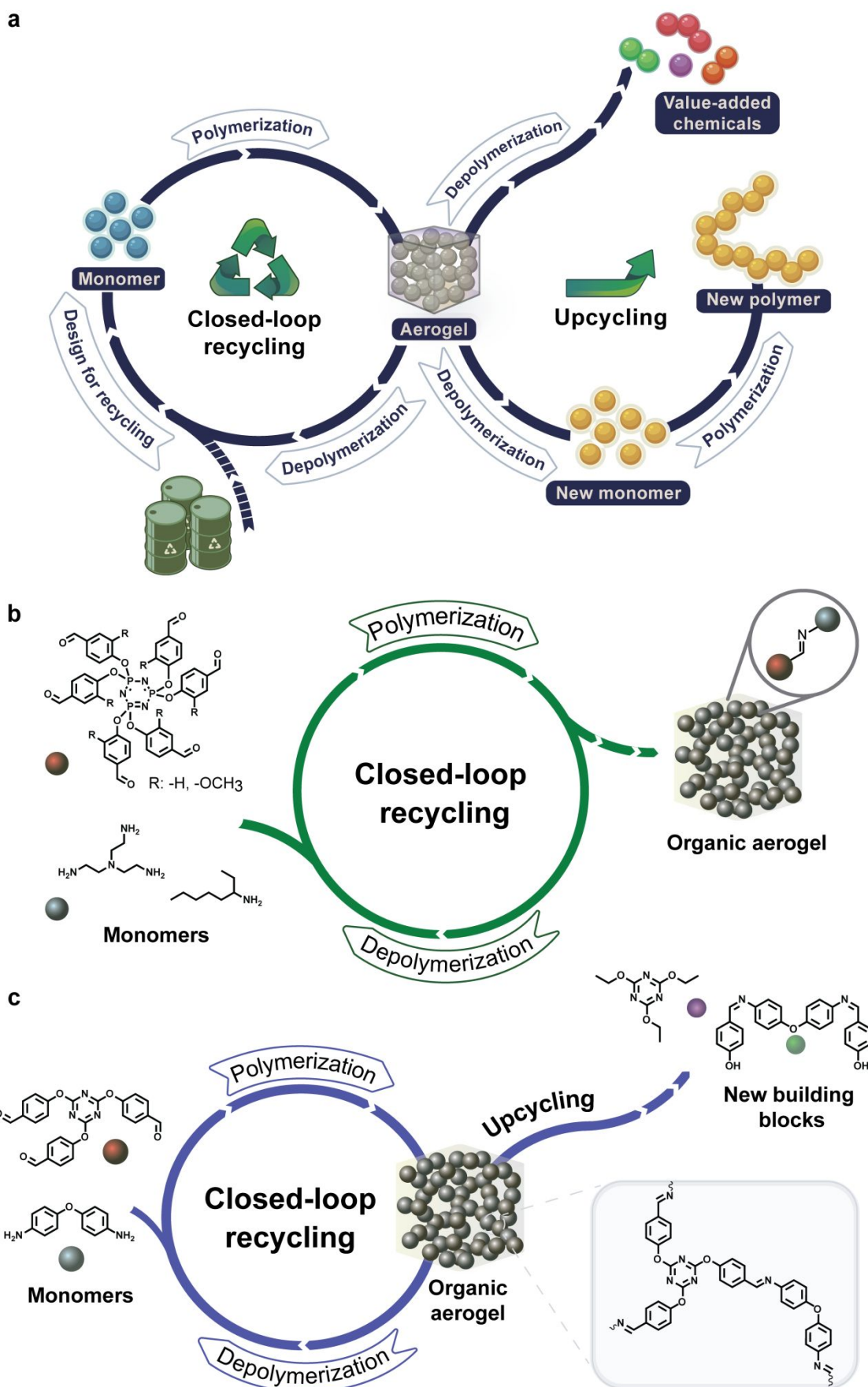
501 Another example of reusable aerogels with little treatment before being reused is α- or β-
502 cyclodextrin-based polyurethane (CDPU) aerogels.¹¹⁴ Upon exposure to a high-humidity (99%)
503 environment at room temperature, these aerogels showed high water vapor absorption
504 capacities (up to 108% w/w). These materials outperformed by far the corresponding
505 cyclodextrins (in powder form) and the commercial products silica gel and Drierite™ (absorption
506 capacities up to 20% w/w) (Figure 5b,c). Most importantly, owing to the balance of enthalpic

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3 507 and entropic factors, absorbed water could be released by just reducing the relative humidity of
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5 508 the environment to 10% at room temperature. CDPU aerogels can be reused for 4-5 times
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7 509 without any significant loss in performance, and they can be reused for at least another 5-6
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9 510 times, as the water vapor uptake seems to have been stabilized, operating at 80% of their
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11 511 maximum performance. This facile regeneration is rather rare and practical and these materials
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14 512 could be used as desiccants in places where cold humid nights alternate with hot dry days.

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17 513 Increasing interest in aerogel production and its applications has raised concern over their end-
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19 514 of-life management. High performance organic aerogels are typically composed of highly
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21 515 covalently cross-linked polymer networks, featuring pronounced chemical stability.³⁷ While this
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23 516 robust design provides organic aerogels with exceptional material properties, it makes them
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25 517 virtually non-recyclable, hindering their end-of-life management. When these aerogels reach
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27 518 the end of their service life cycle, they are either incinerated or disposed in landfills, leading to
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29 519 a loss in resources and a burden for the environment. Additionally, the economic loss of this
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31 520 linear produce-use-discard value chain is substantial and becomes especially important when
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33 521 the starting materials have high value. Reuse of aerogels reduces the burden of manufacturing
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35 522 energy; however, reapplying these materials eventually follows this linear economy model. Due
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37 523 to the lack of effective methods for recycling and valorization of the aerogel waste, valuable
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39 524 resources are lost, and the production of new aerogel materials continues to rely on fossil-based
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41 525 feedstocks and petrochemicals.

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44 526 In stark contrast, the development of fully recyclable aerogels would provide the means for a
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46 527 sustainable circular economy. Therefore, various approaches are being explored to improve the
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48 528 recyclability of aerogels, specifically aiming to achieve closed-loop recycling. Recent ones focus
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50 529 on design of aerogel networks based on non-covalent interactions, including hydrogen
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52 530 bonding,¹¹⁵ electrostatic interactions,^{116,117} and metal coordination¹¹⁸. These non-covalent
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54 531 bonds can be easily broken and reformed under mild conditions, enabling the recycling and
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56 532 reprocessing of aerogels. However, while these reversible interactions offer a potential route to
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3 533 recyclable aerogels, their stability poses a significant challenge. The performance of these
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5 534 aerogels can degrade over time due to environmental factors such as moisture uptake or
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7 535 temperature fluctuations, which can weaken the non-covalent interactions and reduce the
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9 536 material's overall durability, raising questions about their viability for long-term applications.
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12 537 A more promising strategy is the “design for recycling” approach which encompasses the
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14 538 introduction of reversible covalent linkages to fabricate organic aerogels that not only have
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16 539 excellent properties during their useful life but ensures their recyclability under selected
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18 540 conditions (Figure 6a).¹¹⁹ The introduction of such bonds to the aerogel polymeric network
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20 541 facilitates the on demand depolymerization back into original monomers under energy efficient
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22 542 conditions. As the monomers are recovered in high purity and yields, they can immediately be
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24 543 reused to prepare fresh aerogels with identical properties as the original ones (Figure 6b,c).^{120,121}
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26 544 This approach also allows partial depolymerization into soluble oligomers, that can promptly be
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28 545 used to prepare reformed aerogels.¹²² Another key strategy is the incorporation of moieties into
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30 546 the aerogel structure that can selectively be cleaved under specific conditions into value-added
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32 547 chemicals and building blocks (Figure 6c).¹²¹ The design of aerogel structures containing
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34 548 cleavable covalent bonds effectively addresses the environmental and economic challenges of
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36 549 traditional aerogels, paving the way for materials that combine excellent performance with the
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38 550 potential for sustainable, circular aerogel economy (Figure 6).
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3 552 Figure 6. (a) Illustration of the closed-loop recycling and upcycling scheme; (b) closed-loop
4 553 recycling scheme for polyimine aerogels¹²⁰; (c) closed-loop recycling and upcycling scheme for
5 554 polyimine-cyanurate aerogels¹²¹.

6. Process integration strategies in aerogel production

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

6.1. Evaluation of Process Integration Strategies in Aerogels Production

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

565 The combination of standard sol-gel methods with advanced techniques such as supercritical
566 drying can significantly reduce processing time and energy consumption.^{123,124} The use of
567 supercritical CO₂ for the drying phase in aerogel production is a highly efficient and
568 environmentally friendly method. Supercritical drying technique not only reduces solvent
569 residue in the final product to avoid the need for post-processing treatments, but also allows for
570 the recycling of CO₂, minimizing greenhouse gas emissions. Most importantly, this hybrid
571 approach not only preserves the structural integrity of aerogels but also enhances their physical
572 properties. The solvent exchange in gel and supercritical drying steps combined in single
573 apparatus can improve the efficiency of the aerogel production process.¹²⁴ Further on, solvent
574 management should be considered. Instead of using pure solvents, technical solvent mixtures
575 can be applied to improve the economics of the process. However, the effect of the solvent
576 composition on the shrinkage processes and the duration of supercritical drying should carefully
577 be evaluated.¹²⁵

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3 578 The integration of several processes into one, prompted further research on the topic of aerogel
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5 579 production. For instance, the processes of supercritical CO₂ drying and sterilization were
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7 580 integrated into a single one for the production of aerogels suitable for biomedical
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9 581 applications.^{126,127} The integrated process produces decontaminated/sterile and ready-to-use
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11 582 aerogels while reducing processing time without compromising the aerogel's properties for
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13 583 regenerative medicine purposes.

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16 584 Supercritical CO₂ can also be used for impregnation processes that enable the functionalization
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18 585 of aerogels, enhancing the material properties and application potential. *In situ* functionalization
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20 586 can be accomplished using co-precursor techniques or by incorporating functional additives into
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22 587 the sol-gel process.¹²⁸ Recently, the simultaneous starch aerogel formation and curcumin
23
24 588 impregnation were reported to enhance curcumin's bioavailability and storage stability.¹²⁹
25
26 589 Functionalization of aerogels with natural bioactive components can also be performed after
27
28 590 their production, for instance, via supercritical impregnation.¹³⁰ In addition to the impregnation
29
30 591 of neat components, bioactive compounds found in plants can be extracted and impregnated in
31
32 592 aerogels *in situ* using the integrated process of supercritical extraction-impregnation.¹³¹ The
33
34 593 resulting combination of two processes rendered savings in energy and processing time as well
35
36 594 as minimization of extract loss and exposure to air and light. The impregnation processes can
37
38 595 slightly change the morphology of aerogels (i.e., decrease specific surface area because of
39
40 596 precipitation of compounds in pores), but the newly obtained functionalities of aerogels
41
42 597 outbalance this disadvantage. Aerogels can also be used as superior carriers of hydrophobic
43
44 598 synthetic drugs. The thus impregnated aerogels with beclomethasone dipropionate (a
45
46 599 corticosteroid) show excellent aerodynamic properties at relevant doses, as confirmed by *in*
47
48 600 *vitro* lung deposition tests, and the penetration into bronchial tissue as confirmed by *ex vivo*
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50 601 tests with porcine lung tissues.¹³²

51
52 602 The shift from batch to continuous flow processes can streamline production, and reduce waste
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54 603 and energy usage. Continuous flow reactors enable precise control over reaction parameters,
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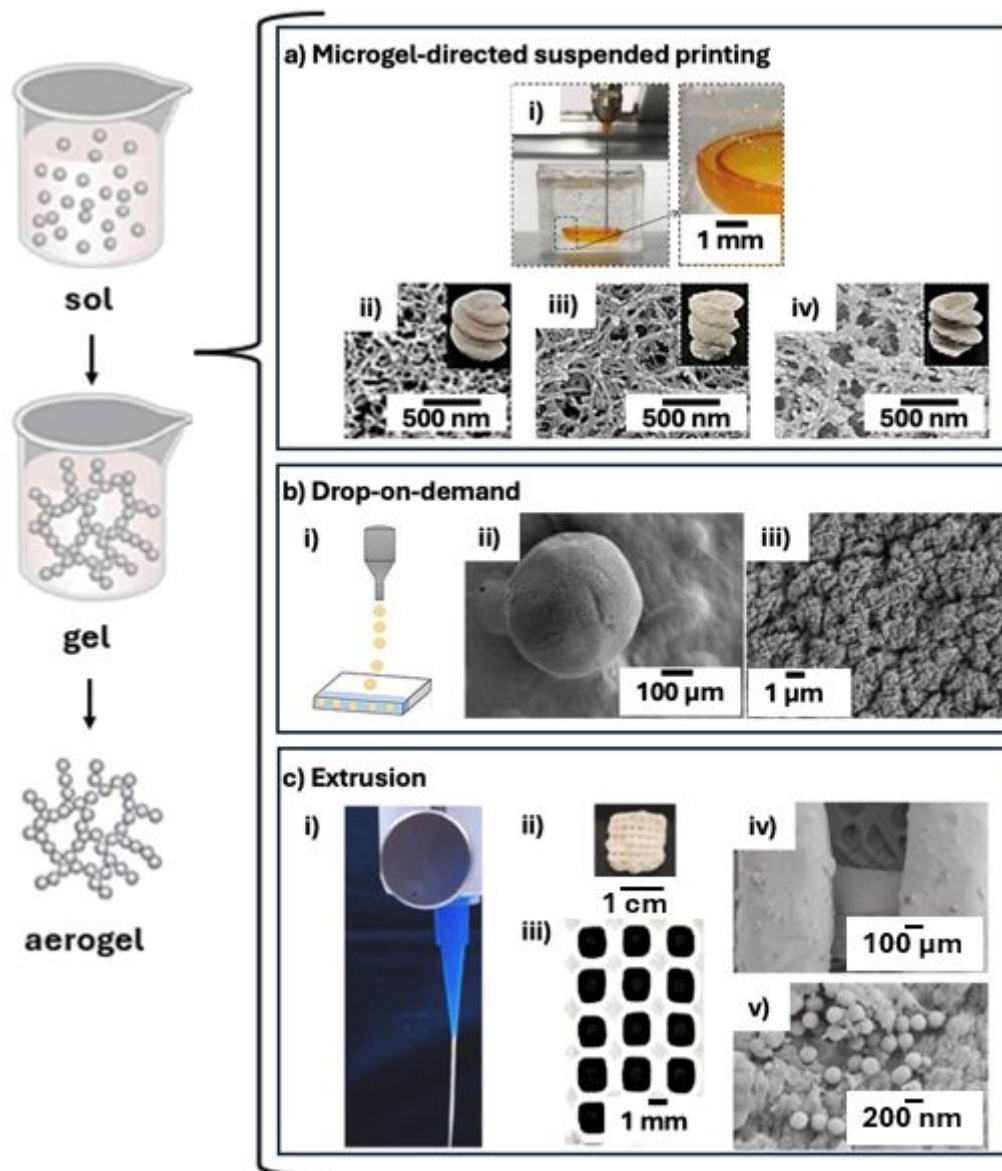
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3 604 leading to uniform aerogel structures and improved scalability. A continuous-mode solvent
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5 605 exchange system can reduce the solvent consumption during the process to one-third with
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7 606 respect to the batch method.¹³³ In addition, the continuous supercritical drying process can
8
9 607 efficiently produce aerogel particles.¹³⁴ A process design involving a counter-current extraction
10
11 608 column with freely sedimenting alginate aerogel particles was proposed using a column of 1.0
12
13 609 m length. The drying of aerogel particles in a shorter column (0.5 m) could be achieved by
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15 610 increasing the CO₂ flow rate, resulting in a 20% reduction in the ethanol outflow mass fraction.
16
17 611 Finally, the integration of energy recovery systems within the production process can capture
18
19 612 and reuse wasted heat, enhancing overall energy efficiency. The integration of industrial waste
20
21 613 heat recovery into smart energy systems represents a main opportunity to accomplish EU
22
23 614 climate and energy objectives.¹³⁵ Such systems can particularly be effective in continuous flow
24
25 615 processes where maintaining optimal reaction temperatures is critical. However, information on
26
27 616 energy recovery system integration within the aerogels production process is still lacking.

31 617 **6.2. Emerging Green Technologies for Aerogel Production**

32 618 Emerging green technologies offer promising avenues for sustainable aerogel production.
33
34 619 Innovative process strategies focus on the rational use of raw materials and energy, including
35
36 620 2D/3D-printing and plasma technology.

37
38 621 Aerogel materials are traditionally prepared using wet sol-gel chemistry, which involves sol
39
40 622 preparation, sol-gel transition, post-treatment, and drying processes.³⁷ Alternative methods
41
42 623 have recently emerged, such as the solid-phase route for perovskite oxide aerogels,¹³⁶ and the
43
44 624 gas-phase route for carbon nanotube (CNT) aerogels¹³⁷. These methods, whether traditional or
45
46 625 newly developed, form the foundation for aerogel manufacturing, including both conventional
47
48 626 and additive manufacturing techniques. Additive manufacturing, particularly 2D- and 3D-
49
50 627 printing, emerged as a cost-effective production technology, suitable for both industrial scale-
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52 628 up and prototyping. This has positioned functional printing at the forefront of the material
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54 629 manufacturing revolution. Unlike traditional material removal, cutting, and assembly processes,
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3 630 printing is a "bottom-up" manufacturing method that builds materials from scratch. It is highly
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5 631 adaptable and potentially more cost-effective than traditional molding methods, allowing for
6
7 632 the production of structurally complex parts that were previously unattainable.¹³⁸
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9
10 633 The first additive manufacturing of aerogels was reported in 2015, utilizing extrusion-based 3D
11
12 634 printing of graphene oxide (GO) inks.¹³⁹ Since then, a wide variety of aerogels, including those
13
14 635 based on graphene,¹⁴⁰ SiO₂,¹⁴¹ resorcinol-formaldehyde (RF) polymer,¹⁴² polyimide,^{143,144}
15
16 636 carbon,¹⁴⁵ cellulose,¹⁴⁶ metal,¹⁴⁷ semiconductor,¹⁴⁸ and g-C₃N₄¹⁴⁹ have been successfully printed.
17
18 637 The technologies employed for printing aerogels include inkjet¹⁵⁰ and microvalve drop-on-
19
20 638 demand printing on water-repellent surfaces¹⁵¹ for microspheres, inkjet and screen for
21
22 639 substrate-bound thin-films,¹⁴⁸ as well as microextrusion,^{152,153} microgel-directed suspended
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24 640 printing¹⁵⁴ and the use of sacrificial templates¹⁵⁵ for creating 3D structures.
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642 Figure 7: Conventional steps in the preparation of an aerogel (left). Integration of 3D-printing
 643 technology into aerogels production with selected examples (right). (a) Microgel-directed
 644 suspended printing set-up (i) Printing of a Kevlar nanofiber in a microgel matrix using such
 645 method,. (ii) cellulose, (iii) alginate and (iv) chitosan aerogels (and their corresponding SEM
 646 images) obtained by using the microgel-directed suspended printing.¹⁵⁴ Reprinted with
 647 permission from ref.¹⁵⁴. Copyright © 2022, ACS Nano; (b) (i) Drop-on-demand printing process
 648 set-up on a superhydrophobic surface,(ii) low and (iii) high magnifications of SEM images of
 649 antibiotic-loaded alginate aerogels microspheres printed by drop on demand.¹⁵¹ Reprinted with
 650 permission from ref.¹⁵¹. Copyright © 2022, MDPI AG; (c) (i) Extrusion-based 3D-printing set-up,
 651 (ii) Visual appearance of 3D-printed alginate aerogels, (iii) 3D-pattern observed on the hydrogel-
 652 based scaffolds. SEM images at (iv) low and (v) high magnifications of upconversion
 653 nanoparticles decorated alginate aerogels.¹⁵³. Reprinted with permission from ref.¹⁵³. Copyright
 654 © 2024, Elsevier.

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3 655 Despite the fabrication method, aerogels are generally fragile due to their low solid content and
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5 656 nanoscale skeletal architecture. However, compared to traditional aerogel shaping methods,
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7 657 "bottom-up" additive manufacturing through functional printing offers unique advantages. In
8
9 658 many cases, these methods allow for the design and shaping of aerogels on a microscale,
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11 659 addressing a significant limitation of traditional sol-gel and mechanical processing methods. The
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13 660 ability to control nanostructures through aerogel chemistry, combined with the macrostructural
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15 661 precision enabled by various printing methods, allows for adaptability and precise control over
16
17 662 aerogel designs from nanometer to centimeter scales. This approach significantly shortens,
18
19 663 simplifies, and improves the processes from basic chemicals or nanomaterials to functional
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21 664 device components, while also reducing production costs. Additionally, additive manufacturing
22
23 665 facilitates the assembly of multi-materials with different functions and structures within
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25 666 different regions of an object,¹⁴⁴ a capability that is typically not possible with traditional
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27 667 fabrication methods (Figure 7). This customization allows to produce aerogels tailored to specific
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29 668 applications, minimizing material waste and enhancing performance. Recently, there has been
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31 669 a growing trend toward using bio-based precursors in 3D printing of aerogels, such as
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33 670 cellulose,¹⁵⁶ alginate,^{157,158} silk fibroin,¹⁵⁹ further enhancing the sustainability of aerogel
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35 671 production in fields like food and medical industries.

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41 672 Plasma technology is another green alternative for aerogel synthesis and surface modification,
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43 673 producing aerogels with unique functionalities. Plasma discharge can facilitate the formation of
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45 674 porous structures and enhance the material surface properties without the need for harsh
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47 675 chemicals. Plasma treatment can be also a fast and versatile technique for deposition of
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49 676 protective hydrophobic and oleophobic polymer layers on hydrophilic biopolymer aerogels. For
50
51 677 instance, hydrophobic modification of biopolymer aerogels (derived from alginate, cellulose,
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53 678 whey protein isolate, and potato protein isolate) was performed by cold plasma coating.¹⁶⁰
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55 679 While the porous structure of aerogels stayed intact during the plasma treatment,
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3 680 polymerization inside the aerogels pore led to the generation of new porous moieties and
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5 681 resulted in a significant increase in the specific surface area.
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10 683 **7. Current consideration on LCA of organic aerogels**

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12 684 LCA is the systematic mapping and evaluation of energy and material flows to critically assess
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14 685 the sustainability of a material or process across the entire life cycle. LCA analyzes the
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16 686 environmental impacts of resource consumption, material production, by-products, waste, and
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18 687 emissions. LCA evaluates a specific design or process in a "cradle-to-gate" or "cradle-to-grave"
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20 688 approach. Obviously, such an analysis is more straightforward for materials that have been
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22 689 applied, or are close to being applied, at an industrial scale. For materials that are still at the
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24 690 laboratory or small pilot scale, many assumptions are required which limit the confidence in the
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26 691 analysis.³⁵
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30 692 According to a 2021 survey of market data,¹⁶¹ over 98% of the aerogel market is composed of
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32 693 silica aerogels, used predominantly for industrial, pipeline and battery thermal insulation and/or
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34 694 protection. Hence, the LCA of silica aerogel production through different synthesis routes has
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36 695 been evaluated, albeit in different frameworks and using different methodologies.¹⁶² Thus, even
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38 696 for silica aerogels, it is therefore not possible to come up with a single measure of environmental
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40 697 impact. However, it is clear that for silica aerogels, the raw materials account for a significant
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42 698 fraction of the embodied emissions.
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46 699 The high impact of raw materials for silica aerogel production made the development of more
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48 700 sustainable, non-toxic, yet cost-effective raw materials a key target of the aerogel scientific
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50 701 community and, to some extent, the aerogel industry. As emphasized throughout this
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52 702 perspective article, the search for sustainability is a key driver for bio-based aerogels, with
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54 703 commonly studied biomass raw materials including biopolymers such as cellulose, alginate,
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56 704 starch, chitosan, gelatin, and whey protein.^{25,27,63,71} However, no bioaerogel products are
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58 705 currently available on the market at industrial scale production volumes, and there are no large-
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3 706 scale production facilities capable of manufacturing bioaerogels in sufficient quantities to meet
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5 707 real-world applications. Consequently, there is an urgent need to consider the LCA of aerogels,
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7 708 as these materials constitute an environmental impact due to their potential production
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9 709 requirements and the existing LCA studies on bioaerogel production are still limited to
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11 710 laboratory-scale analyses. These studies primarily focus on the "cradle-to-gate" scope,
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13 711 sometimes neglecting downstream processes such as utilization and end-of-life (EoL) stages (i.e.
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15 712 product disposal, solvent use and recycling, chemical recycling, and energy consumption).^{35,163–}
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19 713 ¹⁶⁸

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21 714 Although we are not yet able to evaluate the real industrial LCA of biopolymer aerogels, certain
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23 715 aspects can be predicted based on previous studies with biomass processing with other
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25 716 technologies, at small scale for these aerogels or with silica aerogel in industrial production.
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27 717 These studies remain a critical guide for the future development of biopolymer aerogels.
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29 718 Although biopolymer raw materials are inherently more sustainable, extraction methods,
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31 719 modifications, compounding processes, and drying methods can significantly influence their
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33 720 overall environmental impact. As an example, corn cultivation to obtain starch causes a high
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35 721 marine eutrophication (MEP) (54.9%), a high terrestrial ecotoxicity (ET) (50.5%) and a high land
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37 722 occupation (40%).³⁵ Furthermore, although the most part of the needed chemicals during the
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39 723 fabrication of aerogels do not remain in the end product, the way in which they are handled is
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41 724 critical for minimizing environmental effects, especially during the stages of solvent exchange
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43 725 and aging steps, where large amounts of solvents are used. Ethanol is one of the most utilized
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45 726 solvents and therefore, it can represent *ca.* 50% of the global warming potential (GWP) in all
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47 727 types of aerogels and is one of the main contributors to abiotic depletion potential (ADP) and
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49 728 photochemical oxidation.¹⁶⁹ Methanol is also commonly used, and by reducing the number of
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51 729 solvent exchanges carbon emissions can be reduced by up to sevenfold.¹⁷⁰ The drying process,
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53 730 in particular, remains a major challenge for LCA evaluations of aerogels because its significant
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55 731 energy and solvent demands strongly depend on the specific implementation, e.g., on the
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3 732 detailed equipment and process engineering. LCA of the drying step in commercial silica aerogel
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5 733 production provides insights for future optimizations in other aerogel sources. The drying
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7 734 process is both a challenge and a defining feature for aerogels, as it significantly influences their
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9 735 properties and environmental impact. Different drying methods have varying environmental
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11 736 impacts. Freeze drying, commonly used for biopolymer aerogels, is known for its very high
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13 737 energy consumption.^{162,168,171} Ambient pressure drying and supercritical drying involve
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15 738 substantial solvent use due to the close correlation between drying processes and solvent types,
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17 739 leading to both high energy consumption and environmental concerns.^{172,173} Recent studies have
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19 740 highlighted the need for enhancing the sustainability of the production through processes that
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21 741 minimize, eliminate, or recycle solvents and CO₂ (*ca.* 95% recycling rates can be obtained)
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23 742 without compromising aerogel properties.^{35,166,174} Furthermore, optimizing energy consumption
24
25 743 in fabrication processes and utilizing renewable energy sources can greatly reduce the carbon
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27 744 footprint.
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32 745 The environmental repercussions of aerogels can be evaluated through six parameters: GWP,
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34 746 acidification potential (AP), ADP, eutrophication potential (EP), ozone depletion potential (ODP)
35
36 747 and photochemical ozone creation potential (POCP).¹⁷⁵ The transition from lab-scale aerogels to
37
38 748 pilot and industrial scales would reduce in a big proportion the environmental impacts. For
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40 749 example, in the scale-up production of starch aerogels from lab-scale to pilot, the GWP and AP
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42 750 reductions would be of 72%, while from lab to industrial it would be of 95%.¹⁶⁶ The reductions
43
44 751 in EP would be of 61 and 93% for pilot and industrial, respectively; for ODP of 81 and 96%; the
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46 752 electricity use would be reduced in 89 and 99% and the primary energy demand (PED) in a 74
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48 753 and 95%.
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52
53 754 Aerogel waste management introduces further uncertainties in LCA. Biopolymer aerogels,
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55 755 primarily composed of polysaccharides or proteins, are generally considered non-toxic and
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57 756 biodegradable. However, their disposal can be complicated by the inclusion of inorganic or
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59 757 organic crosslinking agents (e.g., chitosan) or surface modifications (e.g., hydrophobization).
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3 758 Composite structures further add to this complexity, and there is currently a lack of
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5 759 comprehensive information regarding their disposal methods and long-term environmental
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7 760 impacts. Finally, tailored LCAs are required for different application scenarios, from thermal
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9 761 insulation to environmental remediation and biomedicine, each with distinct environmental
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11 762 impacts. Addressing these challenges will be crucial for advancing biopolymer aerogels toward
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13 763 sustainable large-scale production and real-world applications.
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17 764

18 19 765 **8. Conclusions, future directions and final remarks**

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21 766 Despite the efforts to revalue waste materials towards innovative uses, aerogels produced
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23 767 directly from waste is still in its early stages. It is important that circularity not only focuses on
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25 768 waste recycling and reuse, but also considers that the processes for aerogels manufacturing or
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27 769 post-modification must be sustainable and scalable, with products being ideally reusable or
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29 770 recyclable. LCA for the production at all levels (from waste collection to end-of-life
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31 771 disposal/reuse of the developed materials) must be done by an economic analysis of the
32
33 772 environmental impact considering the entire process.¹⁷⁶ Economic viability cannot be ensured
34
35 773 solely by defining aerogels as high-value materials, but specific indicators (e.g., financial rate of
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37 774 return to companies and society, benefit-cost ratio) should be evaluated. Also, the alignment
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39 775 with specific United Nations SDGs should be reviewed.¹⁷⁷ In summary, there is still a research
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41 776 gap on the sustainable production of aerogels, which will motivate researchers to develop future
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43 777 aerogel-based materials.
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48 778 Aerogels based on natural resources/waste fractions have some common challenges related to
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50 779 the large variations in the quality and composition of the raw materials. Even when the same
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52 780 biomass is used, this does not guarantee consistent biomass composition, as it is susceptible to
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54 781 significant variations depending on various factors, such as the climate history. Consequently,
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56 782 the properties of the fractions and final products (e.g., color, mechanical and chemical stability)
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58 783 may vary, potentially affecting the process. As an example, impurities originating from plant
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3 784 proteins may lead to catalysts deactivation during initial biomass processing.¹⁷⁸ Consequently,
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5 785 the aerogel production process must be continuously adjusted to accommodate the varying raw
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7 786 material quality, which is currently not considered in process analysis and LCA. In order to tackle
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9
10 787 this problem, stronger involvement of high-resolution climate modelling in the planning of
11
12 788 material production processes is required. Recently, the term “climate-informed engineering”
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14 789 was suggested to address this issue¹⁷⁹ by training a new generation of engineers, who consider
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16 790 climate information in their engineering services similar to the way the economic aspects are
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18 791 considered in their products. Given that bio-based aerogel production combines numerous
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21 792 aspects of material science, engineering and chemistry relevant all over the world, it can serve
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23 793 as a demonstration field for this timely initiative and thus also improve the process
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25 794 sustainability.

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28 795 On the other hand, the evaluation of the human and environmental impact will help to advance
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30 796 in the development and improvement of aerogels to address new health and environmental
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32 797 challenges within the context of the circular economy while complying with the *One Health*
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34 798 concept established by the World Health Organization.¹⁸⁰ The ecotoxicity and health risk
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36 799 assessment of aerogels are not specifically regulated, as aerogels do not require registration as
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38
39 800 nanoforms. Nevertheless, their nanostructured design can raise concerns about a possible
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41 801 hazard assessment, which needs to be addressed. For regulatory purposes, several aspects
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43 802 needed to be considered. As the toxicity inherent to aerogel exposure is not expected in general,
44
45 803 the bioactivity of inhalable or ingestible fragments due to their high inner surface area can raise
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47 804 concerns.¹⁸¹ The pulmonary route due to inhalation of aerogel nanoparticles as material dust
48
49 805 and the consequent pulmonary deposition¹⁸² is the main route of human exposure to the
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52 806 eventual toxicity of aerogels, since fine particles with diameter smaller than 2.5 μm penetrate
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54 807 the alveoli and even reach the cardiovascular system when smaller than 0.1 μm . Professional
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56 808 exposure during the installation and removal activities of insulation materials is one of the most
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59 809 frequent applications where humans are exposed. The use of aerogels on an industrial scale
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3 810 requires the implementation of safety regulations for workers involved in their production and
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5 811 exposure to these nanostructures. Moreover, these particles can be dispersed into the
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7 812 surrounding environment and can circulate in air, soil, and water. Thus, global regulation of their
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9 813 ecotoxicity is needed to prevent any risks to the health of the biosphere and to limit their
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11 814 associated pollution.^{183,184} Additionally, appropriate safety regulations are involved in the
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13 815 manufacturing of aerogel products, identifying hazards for the environment, and human and
14
15 816 animal health.^{185,186} Since aerogels can comprise several chemical compounds, the composition
16
17 817 of each ingredient should be classified as “non-hazardous” in order for the aerogel itself to be
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19 818 considered non-hazardous.¹⁸⁷

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23 819 There are still insufficient studies on the potential toxicity of certain aerogels. A couple of years
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25 820 ago, a systematic toxicological workflow, used to test nanomaterials, was proposed to evaluate
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27 821 and classify aerogels based on their safety profile.¹⁸² Nineteen aerogels, both organic and
28
29 822 inorganic, were compared using a 3 Tier evaluation. The materials’ biosolubility and oxidative
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31 823 potential were tested in Tier 1, the material's toxicity in alveolar macrophages *in vitro* in Tier 2
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33 824 and intratracheal instillation in Wistar rats in Tier 3. All aerogels showed good biocompatibility,
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35 825 except for the case of polyurethane aerogels where a low toxicity potential was detected in Tier
36
37 826 2. From all tested aerogels, only a moderate, transient and reversible inflammation in the lung
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39 827 was found for polyurethane aerogels in Tier 3. In addition to these basic toxicity tests, it is
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41 828 mandatory to evaluate long-term exposition, repeated administration evaluation, bio-
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43 829 absorption, distribution, metabolism and excretion to ensure safe use in biomedical
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45 830 applications.^{188,189}

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49 831 From a data mining perspective, leveraging AI and machine learning algorithms can optimize
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51 832 production parameters, predict process outcomes, and reduce resource consumption. AI-driven
52
53 833 models can identify the most efficient pathways for aerogel synthesis, minimizing the use of raw
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55 834 materials and energy. This direction has been successfully used for aerogel production and
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57 835 application optimization. For instance, it was shown that deep reinforcement learning (DRL) with
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3 836 the diffusion-limited cluster–cluster aggregation (DLCA) algorithm can be applied for
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5 837 microstructural optimization of silica aerogels.¹⁹⁰ Machine learning-based multi-objective
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7 838 optimization using NSGA-II-study of modelling has been applied for an aerogel glazing system in
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9 839 the subtropical climate in order to minimize the total heat gain and maximize the indoor
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11 840 illuminance transmitted through the system.¹⁹¹ Moreover, an artificial neural network (ANN)
12
13 841 was developed for predicting the fractal properties of silica aerogels, given the input parameters
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15 842 for a DLCA algorithm. This approach of machine learning replaces the necessity of first
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17 843 generating the DLCA structures and then simulating and characterizing their fractal
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19 844 properties.^{191,192} Collaborative robotics were also tested in combination with machine learning
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21 845 tools to accelerate the design of conductive aerogels from mixtures of $Ti_3C_2T_x$ MXene, cellulose
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23 846 and gelatine with programmable properties.¹⁹³
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30 848 The visibility of aerogel-based materials and their impact as a top emerging technology have
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32 849 dramatically increased in the last decade. Overall, organic aerogels are multi-functional
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34 850 materials that can be obtained from synthetic or natural materials and have a wide range of
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36 851 applications. The industrial sectors for waste collection (food, textiles, cosmetics, cattle),
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38 852 biorefinery (lignocellulose production), and applications (construction, biomedicine, pollution
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40 853 remediation, critical raw materials recovery, food) are the target groups for expanding the
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42 854 sustainable production and use of aerogels. The awareness and possibilities of these advanced
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44 855 materials can be boosted by new and ongoing international initiatives among the aerogel
45
46 856 scientific community. These initiatives include the implementation of an international
47
48 857 association on aerogels looking for a harmonized voice for the scientific community, the general
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50 858 public and other stakeholders; and to increase networking, training, and other outreach
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52 859 activities. A redefinition of the aerogel term by the IUPAC Association was urgent and has been
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54 860 recently deployed to meet a consensus aiming at limiting the array of materials falling within
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56 861 this material category.
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3 862 The snapshot on aerogel technology provided in this perspective article unveils a promising
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5 863 present and near-future market outlooks alongside an active and growing scientific community.
6
7 864 The glimpse of mid-to-long-term future trends on aerogels technology and applications herein
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9 865 provided is optimistic with novel uses and fast-growing market shares. It also highlights
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11 866 prominent research to adapt conventional aerogel production towards the paradigms of circular
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13 867 economy, and raw material and energy efficiencies. The sustainability principle must be
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15 868 definitely tackled within the aerogel community. Venues for progress and current gaps to be
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17 869 filled in aerogel technology are identified and intense research efforts are needed from different
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19 870 approaches and domains. There are already incipient international collaborative efforts on
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21 871 research, training,^{29,194} and events specifically focused on the cross-fertilization of ideas on this
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23 872 environmental aspect for aerogels, as well as innovation grants funded by public funding
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25 873 bodies^{195,196} to boost business plan initiatives with efficient communication between innovators,
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27 874 industrial players, and business developers.
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34 876 **Biographies**35
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53 878 Prof. Carlos A. García-González received a PhD in Process Engineering in 2009 from the Technical
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55 879 University of Catalonia (Spain), complementing his education during a two-year postdoctoral
56
57 880 stay at the Hamburg University of Technology (Germany) and a two-year industrial experience
58
59 881 at the company Solvay (Brussels, Belgium). In 2014, he moved to the Department of
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3 882 Pharmacology, Pharmacy and Pharmaceutical Technology of the University of Santiago de
4 883 Compostela (Spain). His research within the AerogelsLab team (aerogelslab.egd.gal) is focused
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6 884 on the development of novel processing approaches and sustainable solutions using green
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8 885 technologies to obtain nanostructured materials validated for biomedical purposes. He places
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10 886 particular emphasis on aerogel-based materials intended for drug delivery, wound healing and
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12 887 regenerative medicine.



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24
25 889 María Blanco-Vales graduated in Chemistry from the University of Santiago de Compostela. She
26
27 890 is currently working on her PhD thesis at the same university on technological innovations in
28
29 891 biomedical materials towards their efficient and sustainable production. She is particularly
30
31 892 focused on the reprocessing of biopolymer-based aerogels and on the production of aerogels
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33 893 using different sources of waste as precursors.



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49 895 Joana Barros (JB) earned a PhD (Cum Laude) in Biomedical Engineering at the Faculty of
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51 896 Engineering of the University of Porto (FEUP), in collaboration with the Institute for Biomedical
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53 897 Engineering (INEB)/Institute for Research and Innovation in Health (i3S), and Univ. Minho (UM).
54
55 898 She is currently working as a Junior Researcher (CEECIND, 5th edition) in i3S's Bioengineering
56
57 899 Surfaces Group. Her research has focused on the development of multifunctional approaches
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59 900 based on anti-infective biomaterials and drug delivery systems as biological and bioactive
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901 platforms for effective human infection treatment.

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903 Dr A. C. Boccia received her PhD degree in chemistry in 2004 from the University of Salerno.
904 From 2004 to 2005, she worked as a researcher at Basell Polyolefins in Italy and France. Since
905 2006, she has been conducting research at the CNR-National Research Council in Milan. Her
906 research is focused on the synthesis and microstructural characterization of plastic polymers for
907 different applications, by NMR spectroscopy. Recently, she has broadened her research
908 interests to the development of natural polymers from polysaccharides, particularly for the
909 production of cryogels used in life sciences and packaging. Additionally, Dr. Boccia is contributing
910 to the field of cultural heritage, applying NMR spectroscopy to investigate the degradation
911 mechanisms of plastic artworks and developing strategies for their preservation.

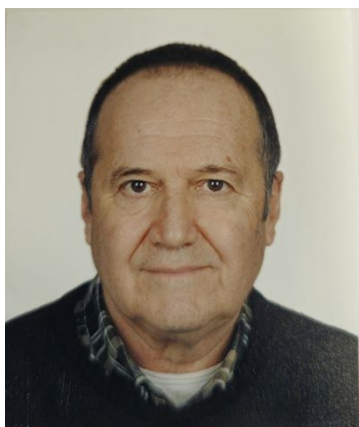
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913 Tatiana is an expert in polymer chemical physics, in particular, in polymer solutions, gels and
914 aerogels, with a focus on biomass-based polymers, and also in polymer composites with natural
915 fibers. She defended her PhD at the Institute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



921

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



929

930 Prof. Dr. Can Erkey received his B.S. in Chemical Engineering from Boğaziçi University in 1984
931 and his Ph.D. in Chemical Engineering from Texas A&M University in 1989. He started his
932 academic career at the Chemical Engineering Department of the University of Connecticut in
933 1995 as an assistant professor. He then joined the Chemical and Biological Engineering
934 Department at Koç University in 2006. His research interests are in hydrogen technologies,
935 nanostructured materials and supercritical fluids. He has been working in the field of aerogels
936 since 2003 with an emphasis on electrocatalytic and adsorptive applications related to hydrogen
937 fuel cells, electrolyzers and gas separation systems.



938

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



947

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

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17 953 Dr. József Kalmár earned his PhD in 2013 at the University of Debrecen, where he is now an
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19 954 associate professor. He established a new research direction focusing on exploring application-
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21 955 related structure-properties-function relationships in porous nanostructured materials. His
22
23 956 team has described the intimate mechanisms of hydration-induced structural changes in
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25 957 aerogels, sorption equilibria, catalytic reactions, and surface complexation of metal ions. His
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27 958 novel mechanistic approach has been utilized in several joint EU projects.

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42 960 Ana Iglesias-Mejuto, PhD in Pharmaceutical Technology from the University of Santiago de
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44 961 Compostela, completed her PhD Thesis focused on the 3D-printing of bioaerogels for bone tissue
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46 962 engineering applications and also on their evaluation by different physicochemical and biological
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48 963 tests. She is currently working on the development of bioinks for the 3D-printing of hydrogels
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50 964 for regenerative medicine purposes.

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



972

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



982

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



988

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

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17 995 Stoja Milovanovic obtained her master's degree in Biochemical Engineering and Biotechnology
18 996 (in 2010) and her doctorate in Chemical Engineering (in 2015) from the Faculty of Technology
19 997 and Metallurgy (University of Belgrade, Serbia) where she currently works as a Senior Research
20 998 Associate. Her work primarily focuses on supercritical carbon dioxide-assisted processes, such
21 999 as extraction from plant material, as well as polymer drying, foaming, and impregnation, to
22 1000 develop new or improved products and materials. She places particular emphasis on sustainable
23 1001 and green processing methods tailored for the pharmaceutical, food, and medical sectors.

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44 1003 Prof. Monica Neagu received her PhD degree in 1996 from the Romanian Academy of Science.
45 1004 She is currently the Head of Immunology Laboratory "Victor Babes" National Institute of
46 1005 Pathology in Bucharest, Romania, and a Habilitated Professor of Immunology at the University
47 1006 of Bucharest. In recent years, she has been involved in immunotoxicology studies of various
48 1007 compounds to be used in the biomedical domain.



1008

1009 Dr. Loredana E. Nita, is a 1st degree researcher at the “Petru Poni” Institute of Macromolecular
1010 Chemistry since the year 2001. In April 2008 she received her PhD degree in Chemistry. In 2019
1011 she obtained the title of doctoral supervisor (habilitation) and since 2020 she supervises 4 PhD
1012 students. Her activity was performed both from a fundamental as well as an applied standpoint,
1013 using a multidisciplinary approach, allowing her to develop the following research directions: -
1014 obtaining pH and thermo-sensitive hydrogels by adjusting the chemical functionality of the gel
1015 structure through the inclusion of a second interpenetrating network and/or specific entrapped
1016 structures, - obtaining hydrogels with a multi-membrane organization through a multi-stage
1017 gelation process; - obtaining and testing systems that have encapsulated drugs, starting from
1018 advanced functional macromolecular structures made by self - assembling process; - testing the
1019 possibilities for the use of hydrogels as a controlled drug delivery system.



1020

1021 Prof. Patrina Paraskevopoulou received her PhD in Chemistry in 2003 from the National and
1022 Kapodistrian University of Athens (NKUA). From 2006 to 2007, she worked as a postdoctoral
1023 fellow at the Missouri University of Science and Technology (MS&T) in the USA. In 2017, she was
1024 a Visiting Professor at the Hamburg University of Technology (TUHH), Germany, funded by DAAD
1025 through its Research Stays for University Academics and Scientists program. She is currently a
1026 Full Professor in the Department of Chemistry at NKUA, Greece. Her primary research interests

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3 1027 focus on inorganic and hybrid organic/inorganic nanostructured materials, such as metal-doped
4 1028 synthetic polymers, biopolymeric and carbon aerogels, and their potential applications in
5 1029 energy, environmental remediation, catalysis, and biomedicine.
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19 1030
20 1031 Prof. Anna Roig holds a degree in Physics and received a PhD in Materials Science from the
21 1032 Universitat Autònoma de Barcelona (UAB), complementing her education at the Royal Institute
22 1033 of Technology in Stockholm and Northeastern University in Boston. Currently, she works at the
23 1034 Materials Science Institute of Barcelona (ICMAB-CSIC), where she leads the Nanoparticles and
24 1035 Nanocomposites Group (nn.icmab.es). Her research pivots around the rational synthesis of
25 1036 nanoparticles and nanocomposites using green chemistry and biotechnology routes while
26 1037 validating those materials for biomedical applications. Specifically, her group has developed
27 1038 nanomaterials as drug delivery vehicles, contrast agents, and bio-based hydrogels/aerogels.
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48
49 1040 Dr. Rosana Simón-Vázquez received her PhD in 2009 from the Autonomous University of
50 1041 Barcelona, Spain. In 2010, she joined the University of Vigo (Spain) as a postdoctoral researcher.
51 1042 From 2014 to 2016, she undertook a research secondment at the Institut Galien, University of
52 1043 Paris Sud (France), and in 2017, at the Center for Research in Molecular Medicine and Chronic
53 1044 Diseases (Santiago de Compostela, Spain). She is currently working as an associate professor at
54 1045 the University of Vigo. Her research interests are primarily focused on nanotoxicology,
55 1046 nanomedicine, and immunotherapy.
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1048 Since 2008 Irina Smirnova is a Professor at the Hamburg University of Technology, Hamburg,
1049 Germany. She serves as the Head of the Institute of Thermal Separation Processes and as the
1050 Vice-president of research of the TUHH. Her research interests include aerogels, high pressure
1051 processes, thermodynamics and separation technologies. She has published over 200 papers in
1052 these fields (<https://orcid.org/0000-0003-4503-4039>). Before that, the engineer was a visiting
1053 scientist at Sogang University in South Korea and worked until 2008 as a group leader and
1054 postdoctoral candidate at the Institute for Thermodynamics and Thermal Process Engineering
1055 at the Technical University of Berlin and at the Institute for Thermal Process Engineering at the
1056 University of Erlangen-Nuremberg. Smirnova studied physical chemistry at the State University
1057 of St. Petersburg and received her doctorate in 2002 from the Technical University of Berlin on
1058 the synthesis and application of aerogels in process engineering. She completed her
1059 postdoctoral qualification in 2008 at the University of Erlangen-Nuremberg. In her scientific
1060 career, she has also been awarded the DECHEMA Young Academic Award, the Hamburg
1061 Teaching Award and the Ralf Dahrendorf Prize.



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3 1063 Željko Tomović obtained his BSc and MSc degrees in Chemistry at the University of Kragujevac,
4 1064 Serbia. In 2001, he joined the group of Prof. Klaus Müllen at the Max Planck Institute for Polymer
5 1065 Research in Mainz, where he completed his PhD thesis in 2005. After a postdoc in the group of
6 1066 Prof. Bert Meijer at the Eindhoven University of Technology, he joined BASF in 2006, working in
7 1067 the field of polyurethane and carbon-rich materials research. In 2020, he became a full professor
8 1068 at the Eindhoven University of Technology. His research interests center on circular and high-
9 1069 performance polymer materials.



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29
30 1071 Dr. Clara López Iglesias obtained her PhD in Drug Research and Development at the University
31 1072 of Santiago de Compostela in 2020. She then obtained a 3-year postdoctoral fellow Xunta de
32 1073 Galicia, which included 2-year research stay at the Free University of Berlin. Currently, she holds
33 1074 another Xunta de Galicia postdoctoral fellowship and works as a postdoctoral researcher at the
34 1075 University of Santiago de Compostela. Her research interests include aerogels, supercritical
35 1076 fluids, drug delivery, nanomedicine, and biomaterials.

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42 43 1078 **Acknowledgments**

44
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1086 of the manuscript.
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3 1734 **Non-technical synopsis**
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6 1735 Aerogels are lightweight materials well-positioned to support industry with more efficient use
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8 1736 of resources as demand for eco-conscious technologies rises.
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