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DOI: 10.2478/eces-2022-0030 ECOL CHEM ENG S. 2022;29(4):447-462

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HOW CAN HYBRID MATERIALS ENABLE A CIRCULAR ECONOMY?

Abstract: Climate change, critical material shortages and environmental degradation pose an existential threat to the entire world. Immediate action is needed to transform the global economy towards a more circular economy with less intensive use of fossil energy and limited resources and more use of recyclable materials. Recyclable materials and manufacturing techniques will play a critical role in this transformation. Substantial advancements will be needed to achieve a more intelligent materials design to enhance both functionality and enhanced sustainability. The development of hybrid materials combining functionality at macro and nano scales based on organic and inorganic compounds, that are entirely recyclable could be used for tremendous applications. In this mini-review, we provide the reader with recent innovations on hybrid materials for application in water, energy and raw materials sectors. The topic is very modern and after its deep study we propose a creation an international research centre, that would combine the development of hybrid materials with green manufacturing. We have highlighted a framework that would comprise critical themes of the initial research needed. Such a centre would promote sustainable production of materials through intelligent hybridisation and eco-efficient, digital manufacturing and enable a circular economy in the long term. Such activities are strongly supported by current environmental and economical initiatives, like the Green Deal, REPower EU and digital EU initiatives.

Keywords: hybrid nanomaterials, water treatment, green energy, hydrogen storage, hydrogen production, smart manufacturing

Introduction

Climate change, resource consumption and resource-restricted availability together with environmental degradation pose an existential threat to the entire world [1]. The overarching objective of the EU *Green Deal* is for the EU to become the first climate-neutral continent by 2050 [2, 3]. Its main components include a circular economy action plan, sustainable industry and elimination of pollution [2]. In this way, the EU (e.g., with the comprehensive help of the program NetZeroCities led by The European

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Institute of Innovation and Technology-Climate [4]) will transform itself into a modern and competitive *circular economy* that uses resources and energy efficiently and become a leader in new technologies needed for this. This becomes even more important in a new political and economic situation after the Ukrainian crisis. There are substantial pressures on materials and manufacturing, including shortages and escalated costs of critical raw materials, resources and energy. ”The new geopolitical and energy market realities require us to significantly accelerate the transition to clean energy and increase Europe’s energy independence from unreliable suppliers and unstable fossil fuels” [5]. REPowerEU is the European Commission’s plan to make Europe independent of Russian fossil fuels before 2030, in view of the mentioned above Ukrainian crisis. The REPowerEU plan sets out a series of measures to reduce dependence on Russian fossil fuels as quickly as possible, accelerate the ecological transformation and at the same time increase the resilience of the energy system across the EU. The concrete measures include application of green hydrogen, energy savings through more efficient and modern production cycles, clean and ecological industry making significantly more use of renewable natural resources and recycling.

Commission President Ursula von der Leyen stressed the need for Europe to lead the transition to a healthy planet and a new digital world. This twin challenge of green and digital transformation has to go hand-in-hand. It requires, as set out in the European Green Deal, an immediate change of direction towards more sustainable solutions which are resource-efficient, circular and climate-neutral.

Applying the principles of the Green Deal must not undermine the EU’s position in the economic space and jeopardise employment, but it should open up new opportunities to innovate, invest and create new jobs. We are already in a huge transformation regarding the way we fabricate products thanks to the digitisation of manufacturing. This transition is so compelling that it is being called the fourth Industrial Revolution (Industry 4.0) [6]. What is now needed is ”Industry 5.0” to enable a further transition to a more sustainable economy that fits within the constraints of the single planet we all live on [7]. Industry 5.0 was designed as a novel production model, in which a significant emphasis is put on the interactions between humans and machines, i.e., it is focused on delivering customer experience, hyper customisation, returning the manpower to factories as well as responsive and distributed supply chain, which could further facilitate product manufacturing. It has been estimated that green manufacturing could reduce carbon emissions by as much as 86 % [8]. The EU stresses the importance of a full cradle-to-cradle cycle (C2C; life cycle analysis terminology for the description of the full recyclability of the materials, i.e., at the end of their life cycle, they are transformed into new functional products) to Industry 5.0, which in turn is critically connected to the Green Deal. The term *eco-effectiveness* is used to describe our ability to deliver C2C, reduce materials and energy intensity and substitute potentially hazardous substances. The current digitisation revolution in manufacturing strongly enables this fifth ”green industrial revolution” to take place [9]. In conclusion, the transition to a green economy represents a significant opportunity and challenge for European industry, creating markets for clean technologies and products [10].

The recent crises have also shown the European industry’s vulnerability to external influences and its dependence on technological imports, energy sources and raw materials. Only modern (digitalised), environmentally friendly (based on green principles) and energy and raw material-efficient industries will prove competitive and ensure this return [11]. However, the challenge is how to bring a step change in eco-effectiveness about, in

a practical sense, for the products society uses. A great fear of many consumers and producers is that an eco-effective, C2C economy means that *quality must suffer*, and that the standards and performance we expect from products today cannot be maintained. One way to tackle this problem would be to take a completely different approach to make the product, focusing on the materials needed to produce it. Thanks to the development of advanced composite materials with natural and degradable components, the massive development of nanotechnology and, last but not least, the shift in the possibilities of additive technologies, it is now possible to think about the artificial creation of materials or their internal structure [12, 13]. This allows to obtain the desired properties, to replace some classical techniques where waste or inefficient use of (raw) materials must occur by principle with brand new, hybrid materials and, in essence, to start talking about the possibility of ”manufacturing” tailor materials having the desired properties but with enhanced sustainability due to (complete) recyclability [11]. By using tailored hybrid materials with ”*designed in*” functionalities we could maximise both manufacturing efficiency and sustainability in parallel. In addition, this approach ”*designs out*” inefficiencies such as wastage, hazardous components, critical materials, and excessive energy consumption. This is why we can start to talk about functionalised, structure-controlled or per-purpose-optimised green materials.

This overall transformation cannot be achieved without a substantial transformation of the extraction, processing and use of materials to manufacture components e.g., for vehicle technology, mechanical engineering and construction. To ensure a circular economy, it also needs to reduce the use of hazardous inputs, given that chemical pollution is recognised as potentially the next most serious challenge to humankind after climate change [14]. So, e.g., are measures against water contamination the next challenge of the present world?

Another question arises: ***How can we produce components from materials in the future that, on the one hand, conserve resources and the environment, require low energy during production and leave a minimal CO_2 footprint; but, on the other hand, retain their performance to fulfill the required functions, such as strength, durability, reproducibility or lightweight?***

One answer to this question is provided by novel, sustainable hybrid materials and structures. Hybrid materials and structures combine the suitable materials in the right place, contributing to a significant increase in performance compared to 1-material components. For example, an H_2 tank made of carbon composite and thermoplastic inner liner has a weight saving of over 70% compared to a pure steel tank [15]. Hybrid designs made of fibre composites, aluminium and steel have also become indispensable in aviation and automotive engineering. For example, body structures with components made of steel, aluminium and CFRP can be found in many vehicles (Fig. 1a). Fibre composites (especially CFRP) are predestined for components with high uniaxial loads, while isotropic materials (steel or aluminium) are primarily used for complex loads or components with crash relevance. In the aerospace industry, for example, glass fibre reinforced aluminium (GLARE) has become established for use in aircraft fuselages and wings (Fig. 1b). This hybrid material, consisting of many layers, each only a few tenths of a millimeter thick, combines high stiffness with high damage tolerance (the hybrid structure prevents crack growth).

Other applications in which hybrid structures are indispensable are found, for example, in wind turbine rotor blades (Fig. 1c). The high stiffness and hardness of steel makes it ideal as a flange for connecting the rotor blade to the rotor hub. Recent developments

by the Fraunhofer Institutes IMWS and IWU focus on combining 3D-printed individual structures (e.g., for medical or orthopedic applications) with a topology-optimised load-path-compliant reinforcement made of CFRP (Fig. 1d).

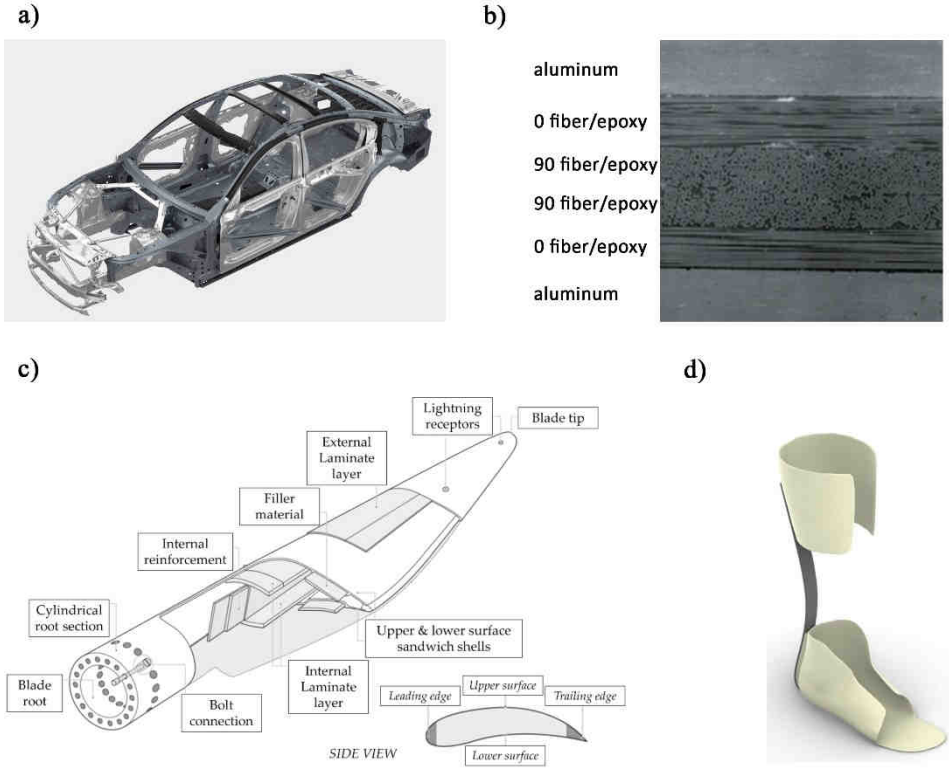


Fig. 1. a) Example of a hybrid body made of steel, aluminium and CFRP used in the BMW 7 Series [16]; b) Cross-sectional view of a hybrid material made of glass fibre-reinforced aluminium (GLARE), which is primarily used in aviation (reprinted with kind permission of MDPI [17]); c) Schematic diagram of a wind turbine rotor blade made of hybrid structure (reprinted with kind permission of MDPI [18]); d) Hybrid ankle orthosis made of patient-adapted 3D-printed plastic shell elements and CFRP stiffening element (Fraunhofer IMWS)

In addition to improving mechanical properties while keeping the component weight low, hybridisation can also be used to specifically tune other functions, such as electromagnetic shielding, tribological resistance or integration of conductive tracks. Hybrid materials and structures are also used in a wide range of micro and even nano applications, such as composites for water treatment [19].

The great advantages of hybrid materials and structures to produce customised, high-performance components have led to a steady spread across many industries in recent decades. In research, too, there have been and still are numerous initiatives and research clusters investigating the material and production technology aspects for implementing hybrid components (e.g., The federal Cluster of Excellence ”MERGE - Technology Fusion for Multifunctional Lightweight Structures” at Chemnitz University of Technology).

However, the major advantages are also offset by a **decisive disadvantage**: The combination of different materials in one component causes **large expenses in the material recycling** of such structures or even prevents them. The material and form-fitting connections between the individual materials are conventionally designed to achieve maximum and long-lasting connection strengths, with the dismantling and separation of the individual materials hardly being taken into account to date. From the point of view of a desired C2C circular economy, hybrid materials and structures require completely new approaches in terms of design, materials and production technology.

Here we propose to make a decisive contribution to this by leveraging the potential of sustainable hybrid materials and digitised green manufacturing technologies to make a solid contribution to environmentally friendly and climate-neutral production and product use and meet the targets of the European Green Deal. This could be achieved by the creation of *an international research centre of excellence* that could produce an exceptional scientific platform for fundamental and applied research projects, which would address the aims of both C2C and Industry 5.0. The focus of IDM2 is to develop fully recyclable hybrid materials and structures and consider new design concepts, technologies and materials. The research aim is to develop completely new ways of joining technologies that ensure maximum strength during service life and can be easily and fully automated disassembled into their individual components at the end of their life cycle without consuming large amounts of energy. Such new joining systems combine both advantages, optimum utilisation of the individual materials according to the principle of ”the right material in the right place” and sustainable recycling of the individual materials into the material cycle. In this centre, we would focus on various technological readiness levels starting from bench scale, through pilot-scale and demonstration. Such a project would exploit innovative data-driven tools in reaching this aim, supporting the European ”twin transition” linking a green economy and digitalisation. Therein, functionalised hybrid materials based on nano/microstructures aim to alter composite parts’ ratio, structure, orientation, size, or other properties would be synthesised and enable using raw materials in many of-value applications. The modern hybrid materials combining inorganic and organic (natural) parts must be exploited in the near future for various applications. These natural-hybrid materials have enormous applications in replacement oil-based products (for example, plastic packaging materials, disposable containers and wraps) [20]. Moreover, hybrid materials can be efficiently used for water and wastewater treatment and purification [19, 21-23], and could be possibly used for cleaning the wastewater that the proposed centre would generate. Furthermore, the energy harvesting and storing by such materials is another future application. For example, as discussed in the following sections, such hybrid materials can be employed for hydrogen storage as well as production. These and other applications will be discussed in future sections; however, first, we will address what hybrid materials exactly are.

What are the hybrid materials and opportunities that they offer?

Standard ”bulk” materials used in industry are often inefficient and composed of toxic and expensive parts, i.e., they are not environmentally friendly. On the other hand, hybrid materials can achieve new functionalities or step changes in performance (e.g., strength to mass ratios) [24].

In this paper, we consider the hybrid materials as components composed of organic (mostly natural and renewable) and inorganic phases [25]. This definition is more prevalent than one of the most common definitions, that the hybrid materials assemblies of nano, micro and macroscale structures, designed from the ”atoms up” [26, 27]. Both of these definitions have in common that such structures should be by some means organised and join two different materials for a specific function.

How to achieve green hybrid materials?

The hybrid materials are the progressive materials of the future, where they fulfil the principles of modern and progressive materials. In order for these materials to fulfil the principles of the Green Deal, circular economy and minimum energy production, it is necessary to maximise their precursors from renewable materials. In other word answer the question ”How can products be made sustainable through intelligent hybridisation and eco-efficient, digital manufacturing?”. We want to propose a framework that would comprise critical themes of the initial research needed. These could include:

- Development of multimaterial compounds that have designed-in C2C functionality using novel interfaces such as switchable adhesives and physical nanostructures (anisotropic physical adhesion) to support re-use / recycling of hybrid materials.
- Inclusion of relevant eco-effectiveness metrics and systematic use of life cycle analysis and carbon footprint in material / product at the research and design stage of products.
- Materials and energy substitutions to reduce resource / energy intensity and reduce use of hazardous substances.
- Smart integration of functionality across scales.
- Effective adoption in materials of recyclates and renewable feedstocks (such as biomaterials) exploiting data-driven & AI based approaches to find resilient strategies to manage variations in material properties.
- Development of tool-free, additive processes as an integral part of in-line production of hybrid parts by printing functional elements (e.g., components, sensors, actuators) onto the basic part structure.
- Development of multifunctional inorganic-organic materials based on designed nano-structure and chemistry and / or based on new 3D techniques in the sub-micron dimension.
- Development of multifunctional bio-degradable coatings and recyclable nano-particles.
- Connecting materials research and engineering with progressive IT (bigdata, artificial intelligence, machine learning), and development of data-driven approach for materials development and manufacturing processes.

Ideally, a research centre, which would offer such possibilities, would also have a digitisation strategy, using the initial seed corn and donated research projects as a platform. The themes of greatest interest would include: a) reducing the staff effort in managing, storing and re-using data and information; b) the development of open source platforms for the exchange and use of data and information; c) the development of operational standards and protocols for data exchange between projects, partners, members and third parties engaging with the centre.

The objective would be to build-up all-embracing databases in terms of material data (production), process data (additive process sequences) and characterisation derived from a well-defined and common meta-data structure and ontologies (such as EMMO) [28].

Digitisation enables the specific design and development of self-improving optimisation of parameters for additive manufacturing processes using machine learning (ML) or artificial intelligence (AI) based principles.

Such a centre would improve three fields of increasing the material, water and energy efficiency (Fig. 2). Several examples of pathways how to achieve this were shown in the subsequent sections.

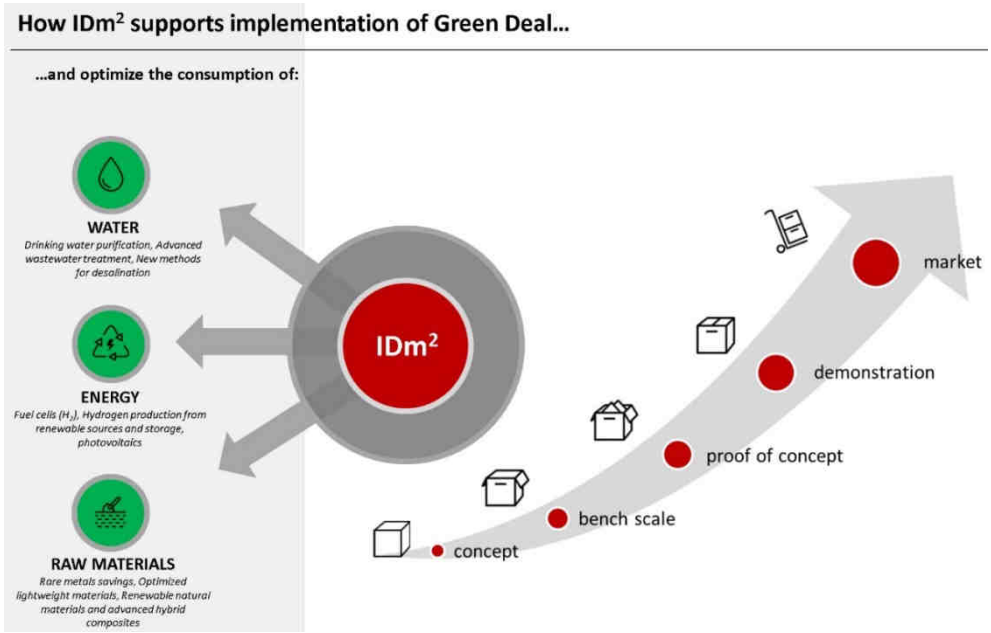


Fig. 2. Scheme presenting the potential of *an international research centre of excellence* (IDm^2) that could produce an exceptional scientific platform for fundamental and applied research projects, which would contribute to the development of raw material, water and energy optimal use

Materials efficiency

In order to produce genuinely sustainable hybrid materials, we should utilise Earth's limited resources in a greener way with the minimalisation of the impact on the environment. Such an approach is in-line with the EU's growth strategy (i.e., resource efficiency [29]).

Hybrid materials can be successfully fabricated from recyclable and renewable sources. Besides long-term used natural and recycled materials, like recycled steel, sheeps wool, wood, straw, cellulose, bamboo, ash, there are new candidates to become fully applicable green materials like recycled plastics, hempcrete, timberecrete, soybean, sugar cane or corn. One example was presented recently as the application of renewable natural materials like natural gums for preparing packaging films and wrapping materials for foodstuff [30-32], which is in line with the conventional oil-based plastics replacement.

Another example of using natural polymers to fabricate hybrid 3D aerogels for the separation of oils and other water contaminants was presented by Ramakrishnan et al. [33]. The material has shown fantastic results for clearing oil spills and as an adsorbent of other

contaminants. Further, such raw polysaccharides can be used to fabricate many other hybrid materials, as presented recently by our group, which synthesised hybrid organic / inorganic composite as gas barriers [34].

Another possible exploitation of raw hybrid materials is as catalysts [35]; for example, red mud, a secondary raw material composed primarily of iron oxides, can be used for pyrolysis and gasification of coal [36] as well as (composed with organic polymers) for many other applications [37]. Moreover, such materials can be utilised for adsorption of various pollutants from water ([38]; for discussion on using hybrid materials for water treatment, please see the next section). Such materials promote the idea of waste-to-product and enable the circular economy.

Concluding, the raw materials can be successfully utilised to fabricate valued hybrid materials. In the authors' opinion, exploiting data-driven & artificial intelligence-based approaches to smoothen the synthesis process set-up can significantly reduce the material and time consumption and should be addressed in further studies for process optimisation.

Water efficiency

Hybrid materials can be successfully used for water and wastewater treatment as filter components [39], catalysts [40] or photocatalysts [41] (which could even be coupled with hydrogen production [42, 43] discussed in the following section).

One example of such an application can be nanometals modified with various organic polymers used for water treatment [44]. For example, by using nano-scale zero-valent iron (nZVI) surface modified with various polymers/organics we can remove multiple contaminants of emerging concern [22, 45-61]. Such hybrid materials have the advantage of rapid pollutant degradation (compared to the biological processes [62]) and often lack of toxic byproducts formation ([63] such as biological removal of perchloroethylene leading to vinylchloride generation). An additional advantage is that such nanocomposites can often be successfully separated by a magnet and reused [64]. Recently, our group has employed supramolecular devices composed of hybrid nZVI-cyclodextrins for target extraction of emerging contaminants from waters, for example, antibiotics [52]. Cyclodextrins, natural materials of variable structure, can sorb various substances into their cavity and in recent years, they have gained an exceptional interest as an adsorbent material for water treatment [65-105]. By the size of the cavity and its polarity (or functional groups), certain substances can be targeted for sorption [19], and they can be incorporated easily into various hybrid materials [19].

In conclusion, hybrid materials have a bright future in water and wastewater treatment; however, creating attempts for their cost-efficient hierarchical production is of high value and should be addressed in future studies.

Energy efficiency

As already mentioned, replacing fossil fuel-based and energy-demanding materials with more eco-friendly materials (bio-based, biodegradable, recyclable) but keeping the same or similar product standards and process quality is one of the primary goals to be achieved.

Hydrogen is one of the greenest renewable fuels; however, there are still significant issues with its efficient production and storage [106]. Photocatalytic production of H_2 from

water as well as its storage for which materials with a very high specific surface area are needed, can be both covered by the fabrication of suitable hybrid nanomaterials.

For hydrogen production, hybrid materials based on covalent organic frameworks (COFs) [107] and metal-organic frameworks (MOFs) [108] can be utilised. Such hybrid materials with appropriate modifications can serve as an excellent photocatalyst for H_2 production. However, further plans on improving the manufacturing and functionalising of such materials should be planned. Primarily to adjust their bandgap to an appropriate range; thus, efficient hydrogen evolution will be achievable under sunlight irradiation [109].

As for hydrogen storage, various hybrid materials can also be effectively used. COF's were reported not only as an efficient material for hydrogen production but also for its storage [110]. Similar to MOF's [111], both benefit from the very high specific surface area, which is very valuable for such applications. More recently, hybrid materials based on electrospun MOF's nanofibers were developed [112], and although the idea of using nanofibers for hydrogen storage is not new [113], many improvements in that 1D material are continuously reported [114, 115]. Moreover, 2D structures such as graphene are among the most promising candidates for H_2 storage [27]. Especially their fabrication with the hierarchical, desired structures could be of value or with various functionalisations [116]. It is expected that such ordered hybrid materials will have a much larger H_2 storage capacity than those available on the market (and in the scientific literature). We also propose to utilise more of the computational approaches for measuring hydrogen adsorption energies on variously functionalised hybrid materials.

Setting a hybrid materials and advanced manufacturing research agenda

A scientific international research centre could provide synergy between hybrid materials and green manufacturing and the importance of data-driven approaches, which could enormously impact scientific/technological, industrial and economic sectors.

The scientific and technological impact of such a project would cover the introduction of new materials and manufacturing technologies that will significantly impact technological processes and the use of more functional and more eco-effective materials and manufacturing to improve existing products and create essential opportunities for new ones. The intention is that these will be competitive with existing products not just on the basis of eco-effectiveness, but also value for money (for example, *via* improved functionality).

The implications for the industrial sector of such a centre would rely on the green and social industrial revolution (Industry 5.0) that the Green Deal will critically depend upon. The new materials and technologies mentioned above can influence production processes in many areas to obtain more eco-friendly production with alternative materials while improving or maintaining their features and keeping the production efficient and economic.

Lastly, the economic impact would base on maintaining or improving product quality in parallel with eco-effectiveness. Although products can be more expensive, the economic return is in the ability to deliver targeted products with longer lifetimes, easier transition to re-use and eco-recycling, which return a significant part of the initial investment. This is seen as likely to have significant wider economic benefits as well as benefitting the industry and business partners adopting these new materials and manufacturing processes.

Concluding remarks

Due to increased anthropogenic actions, there is a need to develop tools that will allow for a sustainable future. Developing novel hybrid materials from renewable sources can become a key to the doorway into a greener future.

As discussed, such materials can be obtained from raw and renewable sources and their selection and fabrication can be enhanced by using advanced artificial intelligence methods. Such hybrid materials can be successfully used for water and wastewater treatment. It is worth mentioning that more hierarchical and ordered structures of such materials can enhance a specific reduction of dangerous persistent organic pollutants, such as perfluorinated alkyl compounds, which are very difficult to remove by conventional (and even to date advanced) processes [117]. Moreover, hybrid materials can be used for obtaining renewable energy (hydrogen) and its storage.

Nevertheless, additional research is needed on such materials' greener and more efficient manufacturing. This goal could be achieved by establishing an international research centre of excellence, which would be focused not only on the fundamental research on greener manufacturing of hybrid materials but also on the transition of their production/use from bench-scale towards the pilot and large-scale applications. Such work would be beneficial for the circular economy programme in the future.

Acknowledgements

The article was created within the call of Horizon - Widera 2022, teaming center IDm^2.

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