Supraparticles for Sustainability

Susanne Wintzheimer, Jakob Reichstein, Philipp Groppe, Andreas Wolf, Bastian Fett, Huanhuan Zhou, Rosa Pujales-Paradela, Franziska Miller, Stephan Müssig, Sarah Wenderoth, and Karl Mandel

The indispensable transformation to a (more) sustainable human society on this planet heavily relies on innovative technologies and advanced materials. The merits of nanoparticles (NPs) in this context are demonstrated widely during the last decades. Yet, it is believed that the impact of particle-based nanomaterials to sustainability can be even further enhanced: taking NPs as building blocks enables the creation of more complex entities, so-called supraparticles (SPs). Due to their evolving phenomena coupling, emergence, and colocalization, SPs enable completely new material functionalities. These new functionalities in SPs can be utilized to render six fields, essential to human life as it is conceived, more sustainable. These fields, selected based on an entropy-rate-related definition of sustainability, are as follows: 1) purification technologies and 2) agricultural delivery systems secure humans “fundamental needs.” 3) Energy storage and conversion, as well as 4) catalysis enable the “basic comfort.” 5) Extending materials lifetime and 6) bringing materials back in use ensure sustaining “modern life comfort.” In this review article, a perspective is provided on why and how the properties of SPs, and not simply properties of individual NPs or conventional bulk materials, may grant attractive alternative pathways in these fields.

1. Introduction

Sustainability is more important than ever to assure future of humankind. In a most simplified definition, sustainability just means the ability to exist constantly. As far as humankind is concerned, it is often defined as living in a way that is not at the expense of future generations. Harmonizing these two demands one can state that the fulfillment of actual human needs (from the very basic to advanced ones), which is required or at least demanded for our continuance of existence, must not sacrifice the well-being of future human generations. All aspects of sustainability may be categorized into environmental, technical, socioethical, and economic concerns, which are all closely interconnected. To reduce complexity, we restrict ourselves to an environmental and technical view in this review on chemical systems. More precisely, we focus on the question of how artificially synthesized objects can contribute to sustainability not only in the environment but also in technical systems and processes that are required to fulfill all human needs in modern life.

This demands to define sustainability in this regard on a fundamental level for which we refer to Lineweaver’s and Townes O'Brien’s article “The Cosmic Context of the Millennium Development Goals: Maximum Entropy and Sustainability.” The authors state that due to the second law of thermodynamics, any system increases in entropy, causing dissipation of energy (into heat) and mater (elements are spread from concentrated reservoirs all over the system, i.e., the planet). However, the sun as major source provides energy to constantly revert this dissipation: With sunlight, biological systems are capable to assemble small molecules into more complex units, biomass is created, and eventually, for instance converted into useful chemical energy reservoirs (e.g., oil). Or, as another example, the sunlight also drives evaporation of water in the oceans, wind—also driven by the sun—carries the moist air to a sink such as a mountain ridge where it accumulates as rain that fills up depleted freshwater reservoirs.

The crucial point is—as the authors outline—that one has to consider the rate of entropy. There is a “sustainable” rate of entropy (dS/dt)sustainable, i.e., a rate at which the “power” provided by the sun can ensure that there is sufficient inversion, keeping the balance between created order and occurring disorder. Hence, for a sustainable living, the rate of entropy on our planet should be minimized.
However, our modern society optimizes processes toward a maximum gain rate and thus artificially increases this rate of entropy generation, and therefore dissipation, that it holds

\[ (\frac{dS}{dt})_{\text{current}} > (\frac{dS}{dt})_{\text{sustainable}} \]  

(1)

Considering once again the second law of thermodynamics, stating that entropy increase is inevitable, one could think that a higher rate of entropy increase may be the natural state every system converges to. However, this is not true as the authors clearly demonstrate that overall, it is decisive for how long entropy in a system can be generated.[4]

Consequently, one has to consider the integral of 1) over time. However, there is a constraint in the integral: if the rate exceeds the chance of a system to “regenerate,” there is a defined point in time \( t \) where the system collapses, see Equation (2).[5]

\[ S_{\text{max}} = \int_{t_{\text{collapse}}}^{t_{\text{sustainable}}} (\frac{dS}{dt})_{\text{sustainable}} \, dt > S_{\text{collapse}} = \int_{0}^{t_{\text{collapse}}} (\frac{dS}{dt})_{\text{current}} \, dt \]  

(2)

A “sustainable” system possesses not only a low rate of entropy increase but also its integral over time can be infinitely extended. This yields a higher overall entropy and thus, fulfilling the second law of thermodynamics better, is the sustainable rate that keeps the ecosystem alive (Equation (3)).[4]

\[ t_{\text{sustainable}} (\frac{dS}{dt})_{\text{sustainable}} > t_{\text{collapse}} (\frac{dS}{dt})_{\text{current}} \]  

(3)

From these considerations follows that an intact ecosystem increases in entropy and thus causes dissipation of any form of resources. However, this proceeds at a rate, where this state can be infinitely extended if an external source of energy is able to renew the condensed reservoirs of resources. Vice versa, a nonintact, thus nonsustainable system is at a state of a too high rate of entropy increase.

Having identified the rate of entropy as the measure for sustainability, one can state that a contributor to sustainability either works against entropy/dissipation increase, using constantly existing forms of energy (sunlight), or helps to decelerate the rate of entropy increase by acting as a storage system, which delays depletion and dissipation.

Based on this, it is possible to identify six fields where materials can contribute to sustainability according to this definition.

1) Purification technologies: As clean water/freshwater is key in our life, reduction of entropy in this system, i.e., inverting water pollution obviously follows the definition. The same holds for air pollution.

2) Agricultural delivery systems: They help to reduce entropy in soil and groundwater, i.e., reduce their pollution.

3) Energy storage and conversion: To convert natural resources such as sunlight into energy we can work with, or transform other forms of energy into stored chemical or electrical energy, obviously helps to create systems that can revert entropy and thus is sustainable according to the definition.

4) Catalysis: Catalysts help to increase the order in a system (e.g., transform several molecules into another, desired, typically higher-ordered compound) and thus, fulfill the definition (less energy is needed to create something of reduced entropy).

5) Extending materials lifetime: Dissipation is decelerated as substances are kept in use for a longer time.

6) Bringing materials back in use: Dissipation is reduced as substances are kept in a cycle of use and reuse.

Going back to the simplified definitions of sustainability mentioned in the very first sentences of this Introduction section, sustainability is the ability to exist constantly due to fulfillment of human needs without sacrificing the well-being of future generations for it. Following this interpretation, the review is structured into the three main sections: “fundamental needs,” “basic comfort,” and “modern life comfort.” Consequently, the identified six fields where materials can contribute to sustainability are classified into these sections. 1) Purification technologies and 2) agricultural delivery systems secure humans “fundamental needs.” 3) Energy storage and conversion, as well as 4) catalysis sustain our “basic comfort.” 5) Extending materials lifetime and 6) bringing materials back in use ensure sustaining “modern life comfort.”

In the last two decades, nanoobjects (nanoparticles) attracted a lot of interest not only due to some unexpected properties that fascinated scientists, but also practically as it turned out that they can significantly contribute to the above listed topics of sustainability in a way that is often superior to bulk materials.[6,7] What all nanoparticles (NPs) have in common is that by transforming the bulk materials to the nanoscale, a) more objects, b) more spatially distributed spots, and c) more surface per gram of material is obtained—with obvious advantages for most application fields. Additionally, true nanoproperties such as quantum effects might emerge.[8]

There were many breakthroughs reported with NPs. Yet, we believe, there is more that can be achieved by going one step further. This next step is to consider the large variety of NPs, tailored in size, shape and composition and therefore in their unique properties, as “the atoms of tomorrows materials”/as the atoms for more complex materials.[9] Just as in supramolecular chemistry,[10] more complex architectures could reveal even more advantageous properties.

Based on existing literature (such as a[11,12] to name but a few), we recently made an attempt[13] to thoroughly define the term supraparticle to include any forms of complex particles made of nanobuilding blocks.[14] Additionally, we outlined, that by combining individual NPs to more complex entities, three new and unique types of properties and thus functionalities can be obtained—which are impossible to achieve with individual NPs. These are coupling, emergence, and colocalization (Scheme 1).[13] Coupling describes the strong interaction between electrons in NPs within a supraparticle (SP) due to their close proximity, while emergence arises from a specific structure within a SP rather than from intrinsic properties of the individual building blocks. Colocalization specifies that different NPs with distinct properties in a SP form a distinguishable common entity with either combined or superimposed properties that can be moved, as well as removed, concentrated, and observed individually. SPs are fundamentally defined as described in Scheme 2:[14] SPs are entities that have been created from colloidally dispersed NPs as starting materials. Processes are excluded where the constituent particles are only transient stages and thus not accessible for analysis, modification, or exchange.[15] SPs are obtained in a dispersed form, for instance, in a colloid, powder, microcomposite, etc. This
distinguishes them from NP arrangements that are infinitely extended in one, two, or three dimensions. Different to randomly agglomerated NPs, a consistent structural motif prevails amongst the obtained SPs, such as crystal packing, fractal dimension, connectivity to the nearest neighbor, defined average size or shape. This results in the creation of additional functionality (coupling, emergence, or colocalization). A more detailed definition and comparison to other terms used for assemblies of NPs, are addressed in an own review article.

In this review article, we point out, where SPs were reported to contribute to “sustainability” according to the definition described in this section. We work out why and how particularly the properties of SPs (and not simply individual NPs) yield unique contributions to the different fields of application.

While in some fields, the development of SPs to achieve superior functionalities is already significantly advanced, some other fields are to date rarely explored. Despite literature is barely available in the latter case, we consider certain fields as especially interesting for SP employment. This is why we included these in the review as a more conceptual part.

2. Supraparticles for Sustainability

As already pointed out in the Introduction, sustainability in a most simple definition means the ability to exist constantly. For living beings first, the basic needs must be secured, namely, water to drink, air to breath, and food to gain energy from. For human beings in a society, second, basic comfort must be sustained to enable mobility, heating, lighting, etc., which all fundamentally depend on electrical energy. Third, our modern world is not only energy but also materials dependent, i.e., “a materials world.”

2.1. Fulfillment of Basic Needs

Guaranteeing the fulfillment of our basic needs not only means to ensure sufficient provision with water, air, and food, but also to secure their cleanliness and thus an appropriate state for consumption. This is why technologies that purify water and air, as well as agricultural delivery systems, reducing unwanted overdosing, contribute to sustainability.

2.1.1. Purification Technologies

One of the most apparent ways to visualize humanity’s (unsustainable) imprint on earth is through pollution of air and water. Since the beginning of the anthropogenic era, human activities have been relentlessly reducing the availability and quality of fresh air and clean water. As both of them are part of the same hydrologic cycle, contamination spreads and makes the imprint of humankind now truly global. Driven by the rapid population growth, fossil fuel-powered economy, and resource-intensive lifestyle, our impact is still accelerating and approaching a critical point with irreversible changes for our planet, like loss of biodiversity and climate change. Not last, human health is directly threatened by the increasing contamination of air and water.

To restore the natural balance and reduce humanity’s imprint, purification technologies will provide a critical contribution to the ultimate goal of a sustainable use of air and water. Most traditional treatment technologies are energy and chemical intensive, as well as ineffective in removing key trace contaminants. For these reasons, there has been a great interest in developing new materials to meet these new needs, especially in the field of nanomaterials.

In the following, we critically discuss benchmark technologies for air and water purification and outline the contribution of specific nanomaterials to provide clean air and water. Based on this, we highlight where the use of SPs yields more sustainable purification technologies.

Air: Air pollution denotes the contamination of indoor and outdoor air by organic and inorganic substances. These pollutants can be classified into three main groups: gaseous pollutants, microorganisms, and particulate matter. Gaseous contaminants include volatile organic compounds, carbon monoxide, nitrogen-containing and sulfur-containing compounds. Microorganisms are bacteria, viruses and fungi and particulate matter describes all solid organic and inorganic pollutants. Air pollution is combated by its prevention or its removal. Most material-based solutions currently focus on the removal of contaminants either from flue gases or from air. However, in order to efficiently eliminate pollutants, they first have to be locally
detected, identified, and quantified. This is why sensors or indicators for the control of air quality are of eminent importance.

Herein, sensor development is mainly concentrated on the monitoring and detection of gaseous contaminants. While there certainly exist many sensor devices based on bulk materials, NPs have also been identified to provide numerous convenient features for their application as sensors and indicators. They offer the ability to synthesize them in almost any type of material, size, or shape and with any chemical surface modification, often showing unique physical, chemical, or electronic nanoderived properties. This permits a precise control of sensitivity and selectivity, as well as response time to physical or chemical parameters. Furthermore, they enable the bottom-up fabrication of flexible sensors due to their small size, which is advantageous over complicated, multistep top-down processes. Finally yet importantly, films with well-controlled porosity can be generated from NPs by precise adjustment of interparticle distances allowing the maximum interaction of a device with its environment. More detailed information about nanosensors can be found in excellent review articles.

Going one step further, using NPs as building blocks to create SPs for gas sensor applications adds several advantages to these materials, while still profiting from the aforementioned features. While NPs easily aggregate during device construction, e.g., during coating fabrication, resulting in a decrease in porosity, SPs maintain their large surface area within their structure (emergence). Furthermore, the micrometer-size of these spheres also provides large pores within coatings, which are needed for the detection of, e.g., large molecular gases, while retaining the preferred (crystal) size of containing NPs (Figure 1a). Based on the colocalization of building blocks, SPs also enable the combination of several materials in close contact, which often improves sensitivity and selectivity or adds supplementary functionalities to the systems.

Several SP-based sensors for the detection of gases like H₂, CO, NO₂, O₂, or NH₃ have already been developed. A highly sensitive H₂S gas sensor consisting of ZnO nanowires decorated with Pd NPs was presented by Zhang et al. (Figure 1b1). The sensor response is measured via the changed resistance in presence of the gas compared to pure air (Figure 1b2). NO₂ can for instance be detected with the help of WO₃ SPs consisting of nanorods also by changes in sensor resistance (Figure 1c). A SP-based support for CO or NH₃ sensitive molecules assembled by differently sized silica NPs was shown by Wintzheimer et al. (Figure 1d1). The gas-sensitive dye was loaded onto the SP after its fabrication and was thus able to operate in dry state instead of only in solution showing, e.g., the presence of NH₃ with a modified reflection intensity (Figure 1d2) and a color shift.

**Scheme 2.** Definition of supraparticles and particles excluded from this definition.

| Supraparticles: | Excluded particles: |
|----------------|---------------------|
| Entity consisting of defined particles created from colloidaly dispersed nanoparticles as the starting material | Constituent particles are only transient stages that are not accessible for analysis, modification, or exchange |
| Are obtained in a dispersed form as colloid, powder, microcomposite, ... | Self-assembled nanoparticle arrangements infinitely extended in 1D, 2D or 3D as fibers, films, colloidal crystals, ... |
| A consistent structural motif prevails among all particles: size, shape or internal structure | Randomly agglomerated nanoparticles |
| Their structural motif creates additional functionality: colocalization, emergence, coupling | |

Adv. Funct. Mater. 2021, 31, 2011089 2011089 (4 of 31) © 2021 The Authors. Advanced Functional Materials published by Wiley-VCH GmbH
Besides other air purification solutions, the current mostly material-based techniques are filtration, adsorption, and photocatalytic purification. Filtration mainly addresses particulate matter, adsorption is most effective for gaseous contaminants and photocatalytic purification cannot only eliminate gaseous contaminants but also microorganisms.\[33]\) While photocatalysis seems to be a promising, powerful air purification tool in which nanomaterials and SPs play an important role, it is discussed in Section 2.2.2. on its own, which is covering SP-based catalysis. Filtration is commonly based on fiber-based mesh or membrane materials, while adsorption traditionally uses activated carbon due to its low cost and effectiveness.\[33\]

Even though these processes may not seem to be suitable for small particles on the first sight, during the last decades the potential of NPs to contribute to this field has been identified. Regarding air filters, NPs can add additional functionality to them, such as antibacterial activity,\[34\] and improve the filter performance either by adding new properties like magnetism,\[35\] or by enhancing an already existing property.\[36\] Due to their large specific surface area, NPs can also provide ultra-high adsorption capacities as adsorbents\[37\] or enable a high loading with adsorbents being used as support material.\[38\]

While still providing very large surface areas due to their inner porosity (emergence),\[39,40\] SPs can provide sizes in the large micrometer (to sometimes even millimeter) scale and

![Figure 1. Scheme comparing the accessibility (arrows) of a1) nanoparticle- and a2) supraparticle-based sensor devices for large gas molecules. Examples for b1–d1) gas sensing supraparticles showing their morphology and b2–d2) their sensor response include ZnO–Pd supraparticles resistively sensing H\(_2\)S (b), WO\(_3\) supraparticles resistively sensing NO\(_2\) (c), as well as a silica-supraparticle-based sensor support for CO or NH\(_3\) sensitive color-changing dyes (d). In all cases (b2–d2), repeated sensing cycles, optionally with increased target gas concentration are depicted. (a1,a2) Adapted with permission.\[23\] Copyright 2014, American Chemical Society. (b1,b2 and c1,c2) Reproduced with permission.\[27,31\] Copyright 2009 and 2016, The Royal Society of Chemistry. (d1,d2) Reproduced with permission.\[32\] Copyright 2019, Wiley-VCH GmbH& Co. KGaA.](image-url)
can thus be directly utilized for gas adsorption processes in common industrially used fixed or fluidized bed reactors.\cite{41,42} Furthermore, their larger sizes compared to NPs also enable a better incorporation without aggregation into filters and a longer sustainment in those.\cite{43}

Ko et al., for example, presented SPs consisting of silica cores and gold NP satellites, which are ideal for the coating of air filter fibers due to their size around 400 nm (Fig. 2a), and proved an additional antibacterial activity of these coated filters (Fig. 2a2).\cite{43} For CO$_2$ capture, MgO SPs were developed providing a large active surface area due to their inner structure and rough surface. Coated with a thin layer of alkali metal salts, they showed a rapid sorption rate and high sorption capacity.\cite{40} Rong et al. created a very pioneering liquid–solid SP by the assembly of hydrophobic silica NPs around a liquid marble (Fig. 2b1), which contained hydrophilic silica NPs assembled with a liquid CO$_2$ absorbing amine. This system exhibited a very high sorption capacity and rate, as well as long-term stability with reduced amine loss in fixed bed reactors (Fig. 2b2).\cite{41} Another SP system addresses the pollution of air by electromagnetic waves, which are emitted by electronic devices and harm human health.\cite{44} Carbon-coated iron oxide NPs (Fe$_3$O$_4$@C) assembled into larger entities efficiently absorb these waves due to their supraparticulate structure (Fig. 2c).\cite{44}

These few examples impressively demonstrate the potential of SPs for the development of improved air purification systems due to their advantageous size range and high surface area.

**Water:** Along with unpolluted air, safe and clean water is mandatory for humankind. It is the origin of life and covers around 70% of the earth’s surface. However, only a very small fraction (0.04%) is accessible for use. Thus, scarcity of clean water is highlighted as one of the biggest challenges of this century.\cite{45} Besides the impact of climate change, inefficient water management, increasing demand, and increasing pollution are accelerating water shortage.\cite{46} Typical, harmful pollutants are specific organic constituents (e.g., pesticides, dyes, pharmaceuticals), heavy metals (e.g., arsenic, lead), high salinity, pathogenic microorganisms and biomolecules, high turbidity from inorganic matter, or a combination of these.\cite{47}

The diversity of pollutants, combined with substantial local differences of water quality across the world, e.g., hardness and alkalinity, makes the purification of water particularly challenging. In order to tackle this, flaws of existing treatment technologies have to be discovered and novel approaches must be developed. More sustainable water purification technologies require a focus not only on improved efficiency but also on other aspects such as a reduced use of primary resources (land, infrastructure), reusability, recyclability, scalability or low costs. Hereafter, we will discuss different important subcategories of water purification technologies: recognition (sensors), disinfection, and decontamination.

Following the goal to detect harmful contaminants in water, the ability to detect water pollution has improved with technological advances,\cite{48} resulting in a broad range of sensors with various signal outputs and application sites within the

---

**Figure 2.** Supraparticles used in air purification systems. a1) Silica-gold supraparticles coated on filter fibers adding antibacterial activity to the system. a2) Their activity was demonstrated against *Escherichia coli* bacteria. b1) Silica-based hydrophilic-hydrophobic liquid-solid amine-containing supraparticles for b2) CO$_2$ sorption in fixed bed reactors. c1) Supraparticles consisting of carbon-coated iron oxide nanoparticles Fe$_3$O$_4$@C for c2) the absorption of electromagnetic waves. (a1,a2) Reproduced with permission.\cite{40} Copyright 2014, The Royal Society of Chemistry. (b1,b2) Reproduced with permission.\cite{41} Copyright 2019, Springer Nature AG & Co. KGaA. (c1,c2) Reproduced with permission.\cite{44} Copyright 2020, Elsevier.
distribution systems.\textsuperscript{[24,49]} Water quality monitoring has evolved manual, off-line control in laboratories to modern online or inline sensors connected via Wi-Fi.\textsuperscript{[50]} The benefits of these novel techniques are a reduction of analysis time and costs, as well as a greatly increased detection rate enabling real-time water control.\textsuperscript{[51]} The implementation of these techniques, however, still has to overcome some barriers. Hence, off-line chromatographic and spectroscopic methods for organic and inorganic chemicals remain the golden standard for water quality monitoring for now.\textsuperscript{[52]}

Nanotechnology-enabled sensors can significantly contribute to the aforementioned transition not only by replacing many existing water quality sensors but also by enabling widespread low-cost measurements over the entire distribution system.\textsuperscript{[49]} This may for instance permit continuous monitoring based on small-scale sensors connected to the Internet of Things.\textsuperscript{[33]} Additionally, nanomaterial-based sensors potentially lead to rapid analysis times and particularly low detection limits enabling the treatment of previously untreatable contaminations.

Although nanosensors have already expanded the horizon of existing technology, we believe that the use of SPs may enable even more sustainable water quality management. Therefore, we highlight some examples of these SP systems in the following section. Plastics are nowadays present in all field of our daily routine. However, there is a growing concern about their release of small toxic molecules, such as Bisphenol A.\textsuperscript{[54]} Uchida et al. designed one promising localized-surface-plasmon-resonance-based SP sensor for the detection of this compound.\textsuperscript{[55]} The SP consists of a polymer NP together with Bisphenol A-modified gold satellite NPs (Figure 3a). While the SP core exhibits imprinted cavities for selective binding of Bisphenol A, the gold NPs act as a signal transducer and due to this core–shell interaction, a new sensor functionality emerges. Whenever free Bisphenol A molecules are present, they competitively bind to the cavities and replace the gold NPs. This results in the disassembly of the SP (Figure 3b) and thus, a signal change is detectable by simple UV-Vis measurements instead of commonly applied enzyme-linked immunosorbent assays. These SPs show outstanding selectivity and enhance the previously reported sensitivity of bulk polymer sensors by a factor of 120 000.\textsuperscript{[55]} Besides introducing new detection pathways, SPs provide an ideal platform to combine multiple functionalities within one complex entity and thereby, improve water sensor performance. Inline nanosensors have the ultimate advantage of continuous surveillance of the water quality due to permanent interaction with the analyte.\textsuperscript{[56]} However, large quantities of sensors are required for sufficient signal intensity and the release of nanosensor materials into the distribution system must be avoided.\textsuperscript{[57]} To overcome this issue, Wehner et al. used the colocalization of two functional building blocks, a super-paramagnetic Fe₃O₄/SiO₂ SP core and different luminescent lanthanide-containing-metal–organic frameworks (MOFs) as the shell.\textsuperscript{[58]} Thereby, target substances can be detected by read-out of the luminescence signal of the MOF-based shell. The magnetic core enables the collection of the MOF@SPs by an external magnetic field and thus, achieves a strong increase of the luminescence signal, while avoiding water pollution due to sensor material release.\textsuperscript{[58]} Similarly, Kim et al. proposed a wash-free sandwich immunoassay using the interaction of magnetic iron oxide and photoluminescent ZnS:Mn SPs.\textsuperscript{[15]} By luminescent detection of the target substance and magnetic harvesting of the sensor particles, they provide a cost-effective method with high sensitivity, specificity, and good reproducibility without time-consuming signal amplification steps. Furthermore, this system is capable of multiplex detection by simply adding a second photoluminescent Cd-doped ZnS SP.\textsuperscript{[35]} Besides continuous surveillance of water quality within the distribution system by inline sensor technology, there is an increasing interest in developing analysis methods for the detection of pollutions in remote areas.\textsuperscript{[49,57]} To develop simple, portable, and low-cost technologies for bioanalysis, smartphone-based solutions are an emerging platform.\textsuperscript{[59]} Tran et al. recently presented a smartphone-based imaging platform for selective cell isolation and quantification and claimed that this technology may also serve as tool for pathogen detection.\textsuperscript{[60]} The key to this technology is an SP system consisting of magnetic iron oxide NPs as core surrounded by photoluminescent quantum dots. The magnetic core enables the isolation of the target for a smartphone-based detection of the optical properties of the SPs.\textsuperscript{[60]} Considering the increasing number of identified harmful contaminants in water, water quality monitoring should focus on multiplex detection of pollutants by semiselective methods like pattern recognition.\textsuperscript{[48,61]} Thereby, selective analyte recognition is achieved through the interaction of an analyte with a sensor array. Subsequently, the combined response is assessed utilizing multivariate statistics resulting in a target selective sensor signal.\textsuperscript{[61]} The development of an SP sensor array based on the colocalization of various selective nanosensors and magnetic NPs would generate a powerful tool for inline and point-of-use water quality monitoring (Figure 3c). This SP could be captured magnetically for signal augmentation and would enable a simultaneous detection of a broad range of harmful and toxic water contaminants, e.g., heavy metal ions, pathogens, and organic pollutants.

The aforementioned systems exemplarily show that SPs have a high potential to contribute significantly to the future of water quality control. They could increase the selectivity and sensitivity of existing sensors, which enables the identification of previously unrecognizable pollutants. Additionally, SPs could facilitate new detection pathways and allow for multiplex detection of contaminants.

The logical consequence of increasing industrial and urban water pollution, as well as the rising global population, is a decreasing available fraction of freshwater per person. In order to fight the accelerating scarcity of usable water, amplified reuse and recycling of wastewater is without alternatives. Besides improved water quality control, this requires advanced purification, i.e., disinfection technologies.

Despite the large improvement in water disinfection technology during the last century, pathogen-contaminated water remains the main cause for waterborne illnesses, particularly in developing countries. The current disinfection technology is based on multiple chemical disinfection steps using chlorine or chloramine and additional germicidal ozone or UV treatments to widen the range of pathogen deactivation.\textsuperscript{[61]} Along with growing concerns about toxic byproducts formed during halogenated disinfection processes, these methods are chemical-, as well as energy-intensive and require extensive infrastructure.\textsuperscript{[62]}

\textsuperscript{[24,49]} Water quality monitoring has evolved manual, off-line control in laboratories to modern online or inline sensors connected via Wi-Fi. The benefits of these novel techniques are a reduction of analysis time and costs, as well as a greatly increased detection rate enabling real-time water control. The implementation of these techniques, however, still has to overcome some barriers. Hence, off-line chromatographic and spectroscopic methods for organic and inorganic chemicals remain the golden standard for water quality monitoring for now.

Nanotechnology-enabled sensors can significantly contribute to the aforementioned transition not only by replacing many existing water quality sensors but also by enabling widespread low-cost measurements over the entire distribution system. This may for instance permit continuous monitoring based on small-scale sensors connected to the Internet of Things. Additionally, nanomaterial-based sensors potentially lead to rapid analysis times and particularly low detection limits enabling the treatment of previously untreatable contaminations.

Although nanosensors have already expanded the horizon of existing technology, we believe that the use of SPs may enable even more sustainable water quality management. Therefore, we highlight some examples of these SP systems in the following section. Plastics are nowadays present in all fields of our daily routine. However, there is a growing concern about their release of small toxic molecules, such as Bisphenol A. Uchida et al. designed one promising localized-surface-plasmon-resonance-based SP sensor for the detection of this compound. The SP consists of a polymer NP together with Bisphenol A-modified gold satellite NPs (Figure 3a). While the SP core exhibits imprinted cavities for selective binding of Bisphenol A, the gold NPs act as a signal transducer and due to this core–shell interaction, a new sensor functionality emerges. Whenever free Bisphenol A molecules are present, they competitively bind to the cavities and replace the gold NPs. This results in the disassembly of the SP (Figure 3b) and thus, a signal change is detectable by simple UV-Vis measurements instead of commonly applied enzyme-linked immunosorbent assays. These SPs show outstanding selectivity and enhance the previously reported sensitivity of bulk polymer sensors by a factor of 120 000. Besides introducing new detection pathways, SPs provide an ideal platform to combine multiple functionalities within one complex entity and thereby, improve water sensor performance. Inline nanosensors have the ultimate advantage of continuous surveillance of the water quality due to permanent interaction with the analyte. However, large quantities of sensors are required for sufficient signal intensity and the release of nanosensor materials into the distribution system must be avoided. To overcome this issue, Wehner et al. used the colocalization of two functional building blocks, a super-paramagnetic Fe₃O₄/SiO₂ SP core and different luminescent lanthanide-containing-metal–organic frameworks (MOFs) as the shell. Thereby, target substances can be detected by read-out of the luminescence signal of the MOF-based shell. The magnetic core enables the collection of the MOF@SPs by an external magnetic field and thus, achieves a strong increase of the luminescence signal, while avoiding water pollution due to sensor material release. Similarly, Kim et al. proposed a wash-free sandwich immunoassay using the interaction of magnetic iron oxide and photoluminescent ZnS:Mn SPs. By luminescent detection of the target substance and magnetic harvesting of the sensor particles, they provide a cost-effective method with high sensitivity, specificity, and good reproducibility without time-consuming signal amplification steps. Furthermore, this system is capable of multiplex detection by simply adding a second photoluminescent Cd-doped ZnS SP. Besides continuous surveillance of water quality within the distribution system by inline sensor technology, there is an increasing interest in developing analysis methods for the detection of pollutions in remote areas. To develop simple, portable, and low-cost technologies for bioanalysis, smartphone-based solutions are an emerging platform. Tran et al. recently presented a smartphone-based imaging platform for selective cell isolation and quantification and claimed that this technology may also serve as tool for pathogen detection. The key to this technology is an SP system consisting of magnetic iron oxide NPs as core surrounded by photoluminescent quantum dots. The magnetic core enables the isolation of the target for a smartphone-based detection of the optical properties of the SPs. Considering the increasing number of identified harmful contaminants in water, water quality monitoring should focus on multiplex detection of pollutants by semiselective methods like pattern recognition. Thereby, selective analyte recognition is achieved through the interaction of an analyte with a sensor array. Subsequently, the combined response is assessed utilizing multivariate statistics resulting in a target selective sensor signal. The development of an SP sensor array based on the colocalization of various selective nanosensors and magnetic NPs would generate a powerful tool for inline and point-of-use water quality monitoring (Figure 3c). This SP could be captured magnetically for signal augmentation and would enable a simultaneous detection of a broad range of harmful and toxic water contaminants, e.g., heavy metal ions, pathogens, and organic pollutants.

The aforementioned systems exemplarily show that SPs have a high potential to contribute significantly to the future of water quality control. They could increase the selectivity and sensitivity of existing sensors, which enables the identification of previously unrecognizable pollutants. Additionally, SPs could facilitate new detection pathways and allow for multiplex detection of contaminants.

The logical consequence of increasing industrial and urban water pollution, as well as the rising global population, is a decreasing available fraction of freshwater per person. In order to fight the accelerating scarcity of usable water, amplified reuse and recycling of wastewater is without alternatives. Besides improved water quality control, this requires advanced purification, i.e., disinfection technologies.

Despite the large improvement in water disinfection technology during the last century, pathogen-contaminated water remains the main cause for waterborne illnesses, particularly in developing countries. The current disinfection technology is based on multiple chemical disinfection steps using chlorine or chloramine and additional germicidal ozone or UV treatments to widen the range of pathogen deactivation. Along with growing concerns about toxic byproducts formed during halogenated disinfection processes, these methods are chemical-, as well as energy-intensive and require extensive infrastructure.
Therefore, advanced affordable disinfection technologies addressing a broad range of pathogens with efficient reuse strategies, reduced energy, and chemical imprint are desired.

Nanomaterials, with their unique properties, such as the often aforementioned large surface area, as well as the specific reactivity and photocatalytic activity, represent a class of material with enormous potential for water disinfection processes.\[63,64\]

Even more effective and thereby, more sustainable water disinfection can result from the assembly of these nanomaterials into SPs. Andra et al. reported that by self-assembly of CuO nanospindles into CuO nanoflowers an enhanced antimicrobial activity compared to isolated nanospindles can be achieved due to coupling effects.\[65\] Moreover, SPs provide an ideal platform for combining different antimicrobial nanomaterials in one disinfection agent, as recently suggested by Mauter et al.\[20\] The colocalization of TiO$_2$, Ag, ZnO NPs, and carbon nanotubes in a single SP would yield a versatile tool with multiple pathogen inactivation pathways for efficient wide-range pathogen deactivation.\[66–69\] In sum, antimicrobial SPs could render water disinfection more effective using simultaneously various inactivation mechanisms, thus, require less energy, chemicals, and infrastructure. However, despite the tremendous potential of SPs for pathogen inactivation, very few examples were reported to date.\[20,65–69\]
Besides the ever-present problem of pathogens, human-derived pollutants in water, such as pesticides, dyes, and pharmaceuticals, have a detrimental effect on human health and ecosystems.[9] Current water decontamination processes include physicochemical separation (adsorption, membrane filtration, and chemical precipitation or coagulation) and chemical oxidation through reactive radicals produced in advanced oxidation processes. These conventional treatment methods lack effectiveness and selectivity, remain chemical- and energy-intensive, and generate harmful chemical byproducts, as well as waste, which requires additional treatment steps, and thus, causing extra costs.[20,71] Therefore, new technologies are demanded that increase cost efficiency, enhance selectivity toward key trace pollutants, require fewer chemicals and less energy while enabling different application areas.

Nanoadsorbents gained tremendous interest as they offer significant improvements over conventional ones, mainly due to their high specific surface area, as it has already been mentioned before, which leads to a high adsorption capacity. Their tunable pore size and their functional surface chemistry enables selective and target specific adsorption of, e.g., toxic heavy metal ions or herbicides.[64,72] Driven by the extensive research effort in this field, nanomaterials with impressive adsorption capacity and highly effective catalytic degradation of pollutants have been developed, to date.

However, through the combination of various NPs in one SP decontamination agent the performance, applicability and sustainability of treatment processes to real water matrices are likely to be further improved.[10,67] Numerous SPs for wastewater decontamination were developed that use enhanced catalytic properties or smart combinations of catalytic and other functional building blocks. As catalysis is a very broad topic, we have devoted Section 2.2.2 of its own to it, where systems and performance for this purpose are described in more detail.

For the design of advanced adsorbent systems, many SPs combine magnetic NPs with other nanomaterials: Sylvester et al. proficiently incorporated small iron oxide NPs into durable highly porous polymer microbeads.[74] These hybrid SPs combine the excellent handling, flow characteristics, and attrition resistance of conventional ion exchange materials with the outstanding affinity of iron oxide NPs for arsenic. Furthermore, several researchers combined the magnetic properties of iron oxide with highly effective adsorbent materials, like zeolitic frameworks[75] or carbon-based nanomaterials,[76] to enable their recovery and reusability. These SPs can be used for efficient decontamination of wastewater from toxic, volatile organic compounds,[77] heavy metal ions,[78] and dyes.[79] To enhance the sustainability of these systems, such compounds can be synthesized via upcycling of agricultural or household waste, like tea waste, and therefore, might be cost-effective adsorbents of the future.[80] After successful magnetic harvesting of adsorbents, multiple regeneration pathways for adsorbents have been proposed. Besides extracting the agents and subsequent reactivation via washing procedures or energy-intensive thermal treatment, additional building blocks are utilized to create more practical and sustainable regeneration methods. For instance, carbon coatings or NPs facilitate effective in situ electrocatalytic or electrochemical regeneration.[81,82] For electrocatalytic denitrification of lake water, Su et al. prepared carbon-coated Fe3O4 SPs via self-assembly of oleic acid capped iron oxide NPs with subsequent carbonization (Figure 4a1). Thereby, a hierarchical structure of Fe3O4 NPs with a rough carbon surface was obtained (Figure 4a2). As a result, these SPs achieved an excellent removal capacity for nitrate (Figure 4a3). Furthermore, the rough carbon surface allowed the electrocatalytic conversion of adsorbed NO3− to N2 and NH4+. Besides recovery granted by colocalization of magnetic NPs and adsorbents, the assembly of NPs to hierarchically structured, porous SPs leads to significant improvements in adsorption capacity and kinetics.[83] Oppmann et al. designed reusable SPs from iron oxide and silica NPs that show a capillary effect due to emergence of the nano building blocks taking up dye from wastewater within 1 min (Figure 4b).[84] By tuning the size of the used silica NPs from 100 to 10 nm the equilibrium time and adsorption capacity can be tailored toward a specific application. Even more sustainable water treatment results from processes designed not only for extraction of contaminants but also for recovery and reuse of valuable substances.[85] In this context, nanoporous sorbents and SPs[86] may provide a smart answer to remove target substances, like phosphate, from water, as recently reviewed by Othman et al.[87] Furthermore, the removal and recovery of heavy metal ions using SPs consisting of superparamagnetic iron oxide NPs confined in a silica matrix has been shown.[88] The captured target substances can be desorbed and enriched in an acidic solution, and thereby, the SPs are regenerated. Adding layered double hydroxide ion exchangers to the above-described SP system, phosphate ions are efficiently adsorbed and magnetically recollected from municipal wastewater in laboratory to pilot-scale applications.[89] To close the loop, the captured phosphate was successfully transformed into struvite, a solid fertilizer product.

As a brief summary of this section, SPs provide a versatile platform (Figure 4c) to render water disinfection and decontamination processes more sustainable by improving their efficiency while reducing chemicals and energy consumption. They further grant novel pathways to provide clean and safe water. Additionally, SPs can be a tool to recover substances from wastewater and keep resources in circulation.

### 2.1.2. Agricultural Delivery Systems

While the preservation of clean air and water definitely is the basis for the long existence of humankind, one should not forget that the provision of food is of the same importance. As nowadays large parts of it are provided to us by agriculture, this section will focus on the potential contribution of SPs to a sustainable agriculture. In this context, sustainability can be understood as the seek for yield increase with the help of biocides, herbicides or fertilizers while trying not to compromise the cleanliness of soil, groundwater, and crop. An approach to address this challenge are delivery systems that provide the needed substance over long term with controlled release rates and minimized uncontrolled dissipation.

Concerning current material solutions, NP-based systems have already been reported by several groups to be applied as biocide, herbicide or fertilizer delivery systems. They have structural advantages like large surface areas, modifiable surface and
stability, and thus superior loading capacity. For example, Gao et al.\textsuperscript{[90]} showed that mesoporous organosilica NPs used for pesticide delivery, have a longer Sclerotinia disease protection window on plants, which provides a solution for precisely managing plant disease and farming. Appell et al. also concluded that nanomaterials can deliver bioactive substances directly to

*Figure 4.* Supraparticles for advanced water disinfection and decontamination. a1) SEM and a2) TEM images of carbon-coated iron oxide supraparticles for electrocatalytic denitrification of water. a3) These supraparticles achieved a nitrate removal capacity of up to 1816 mg N g\(^{-1}\) Fe in 0.1 M Na\(_2\)SO\(_4\) system in 100 mg L\(^{-1}\) nitrate solution within 48 h. SEM images of raspberry-like iron oxide silica supraparticles for rapid and efficient dye uptake: b1) detail, b2) cross-section, b3) cross-section detail, and EDX measurement (inset). c1) Supraparticle consisting of colocalized antimicrobial building blocks, e.g., TiO\(_2\), Ag, ZnO NPs and carbon nanomaterials, as shell and iron oxide nanoparticles as their core for wide-range pathogen deactivation and magnetic recovery; c2) multifunctional magnetic supraparticle for simultaneous disinfection and decontamination to provide safe and clean water in a one-step process that allows for inline and point-of-use water treatment. Such a supraparticle may consist of TiO\(_2\), Ag, and iron oxide NPs, as well as selective adsorbents, e.g., layered double hydroxides (LDHs) and CNTs. (a1–a3) Reproduced with permission.\textsuperscript{[81]} Copyright 2019, American Chemical Society. (b1–b3) Reproduced with permission.\textsuperscript{[84]} Copyright 2018, Wiley-VCH GmbH & Co. KGaA.
target points through controlled release and thus, can reduce the use of agrochemicals.\[91\]

However, over the years, concerns arose about the bioaccumulation of nanomaterials in soil and groundwater and about the resulting unwanted uptake of potentially toxic NPs by humans. This consequently limited their use as agricultural delivery systems.\[92\] Furthermore, NPs are easily taken up by plants and therefore, may even contaminate human food sources.\[93\] Health concerns\[94\] surely not only restrict the application of NPs in agriculture but also in most of the other fields discussed in this review article. The use of SPs, providing larger particle sizes (typically in the microscale), may help to overcome these worries. The handling of any powders containing NPs within SPs reduces dust formation being thus easier and safer.\[95\] Furthermore, SPs are unlikely to cross most biological barriers and are not taken up by most cell types reducing their health hazard.\[96\]

This is also why the use of SPs, as assemblies of NPs, for agricultural delivery systems has a high potential: they still provide the advantages of nanomaterials while increasing the particle size for a safer handling and less material dissipation (Figure 5). A pioneering example of SPs for agricultural delivery systems was presented by Mattos et al.\[97\] Millimetric scaled SPs were assembled by biogenic silica particles and cellulose nanofibrils. Compared to NPs, SPs presented lower mobility, which is associated with the size, and higher UV protection, as the “external” surface area-to-bulk ratio is decreased. Another example regarding the superior properties of SPs was reported by Xie et al. They designed a superhydrophobic biopolymer-coated slow-release fertilizer through self-assembly, which showed extended slow-release longevity (100 days more than unmodified ones) and superior durability in soil environment.\[98\] Additionally, SPs possess a high loading capacity due to their high porosity (emergence) and thus, higher delivery efficiency compared to NP delivery systems.\[99\]

It is also noteworthy that while most other of the SP-based delivery systems found in literature so far are used for medical purposes, their working principle may easily be transferred to agricultural purposes. For instance, Wang et al. investigated mesoporous silica SPs for innerear drug delivery and demonstrated that the concept of using these SPs as carriers is transferable to other delivery systems.\[99\] The creation of SPs may enable the triggered release of substances by blocking the guest-molecule carrying pores of a porous particle with removable NPs. Zhu et al. summarized various controlled-release delivery systems based on mesoporous silica materials and nanoparticulate gatekeepers, working via pH-response, ATP, as well as glutathione- and H$_2$O$_2$-responsive mechanisms.\[100\] Similar to these approaches, Torney et al. loaded a mesoporous silica NP with a gene and its chemical inducer, using gold NPs as caps of the ends, subsequently releasing the chemicals and triggering gene expression via removal of the gold NPs.\[101\] Also NP-raspberry-like containers were reported, which can be applied to carry and release antimicrobials in a controlled manner.\[102\]

All in all, SPs have a promising perspective toward sustainability in the field of agricultural delivery systems. When applied in herbicide/biocide/fertilizer delivery systems, they may help to reduce the use of agrochemicals and thus, decrease damage to water and soil. Additionally, compared to NPs, they are less likely to cause food contamination, as a result of their larger size.

### 2.2. Basic Comfort

Beyond fulfilled fundamental needs, such as clean water, life quality increases with access to (electrical) energy.

#### 2.2.1. Energy Storage and Conversion

The “sustainable” generation of energy, which is the commonly used term for what is the conversion of various energy forms from the renewables wind, water, and sunlight into electrical energy, is heavily driven by materials development. In the context of electrical energy generation, SPs are not yet playing an important role. However, with regard to water and wind as energy sources, SPs may be considered as alternative design approaches for rare-earth element-free permanent magnets in the future.\[103\] For solar cells, SPs seem to be of interest for several types of photovoltaic technologies like perovskite or quantum dot solar cells.\[104\] For instance, Zhou et al. incorporated Cu$_2$S$_3$@SiO$_2$ SPs in a hole transport layer of a perovskite...
solar cell. Besides improving the transport capacity and the electrical conductivity, the strong light scattering and reflection properties of the SPs increased the light trapping within the device, resulting in greater absorption efficiencies.\[105\] Especially in the field of quantum dot solar cells, where a well-defined assembly of quantum dots is an essential criterion to make use of coupling mechanisms, SPs may contribute to the ongoing process of implementing the promising theoretical capacity of the technology into well-performing photovoltaic devices.\[106\] A key issue in quantum dot solar cells is the extraction of photo-excited charge carriers before trapping or recombination. In order to tackle this challenge, Ojha et al. conjugated graphene sheets with SPs composed of CdS quantum dots. This coupling provided more stable excitons as well as ultrafast electron transfer from the photoexcited CdS quantum dots to the graphene sheets, leading to improved charge separation efficiencies.\[107\]

Since generation and usage of electrical energy are usually separated by time and location, intelligent distribution and efficient storage are key factors for a sustainable handling of energy. The further improvement of energy storage devices like batteries, supercapacitors, and fuel cells is therefore inevitable to tackle these challenges on the entire scale, from stabilizing the electric grid down to mobility topics and more sustainable portable electronic devices. In order to enhance the performance of such energy storage devices, the application of SPs has indeed attracted increasing interest in recent years.

In the versatile field of batteries, it is especially the lithium-ion technology that stands out with high energy and power densities. On top of that, the efficient conversion from electrical into chemical energy and vice versa, as well as the extremely low self-discharge rate prevent energy dissipation and guarantee entropy conservation. However, there is still a large gap between the performance of modern lithium-ion batteries and the theoretical potential of the technology. The following section displays how SPs and their unique properties help to reduce this gap.

The electrodes, the key components of every battery, are responsible to convert electrical into chemical energy via a redox reaction accompanied by storage and release of lithium-ions. In order to have both, high energy and power densities, the active materials of the electrodes need to possess high specific capacities, as well as high ionic and electronic conductivities. On top of that, some active materials undergo significant changes in volume while storing and releasing lithium-ions, causing mechanical stress in the electrode.\[108–110\] In order to cope with volume changes and to allow electrolyte wetting, electrodes are commonly designed as porous layers including micrometer-sized active material particles, conductive carbon to improve the electronic conductivity and polymer binders (Figure 6a). However, active materials generally possess poor conductivities and dense micrometer-sized particles still represent long pathways for lithium-ions and electrons, which significantly limits the electrode performance. Thus, the logic step is to go down to the nanoscale, i.e., NPs, to create higher surface areas and shorter pathways (Figure 6b).\[111\] Furthermore, in theory nanoporous structures are able to cope better with volume changes than micrometer-sized particles that will crack under high mechanical stress.\[112\]

Yet, by reducing the particle size to the nanoscale, new challenges arise, including uncontrolled aggregation, increased moisture absorption, and broad particle size distributions that lead to higher demands of binder, as well as drastically

---

**Figure 6.** Superiority of supraparticle employment as active material in lithium-ion batteries. a) General build-up of an electrode layer and the issues of dense micrometer-sized particles. b) Nanoparticles (NPs) and the pros and cons as active material. c) Most common designs for supraparticles and the example of a supraparticle that combines various design approaches.
varying rheological properties of electrode slurries. Liu et al. prepared carbon-coated NPs of the cathode material LiFe$_{0.6}$Mn$_{0.4}$PO$_4$/C (here: positive electrode) and compared them with SPs that were produced by spray-drying of these NPs. Besides the advantages in terms of processing, these particles exhibited an increased tap density that resulted in a significantly higher volumetric energy density. On top of the processing advantages that result from this controlled colocalization, a well-defined design can greatly enhance the electrochemical properties of SPs. By spray-drying an aqueous dispersion of LiFePO$_4$ nanorods with glucose, Yoo et al. synthesized SPs with micro- and mesopores. The combination of carbon coating and the hierarchical 3D structure lead to an outstanding performance at high current densities due to improved conductivities. Two further commonly used design approaches for SPs in batteries are multicomponent composites and the embedment of the NP building blocks in a matrix material (Figure 6c). Since every active material has its unique properties, the design of the SP can be tailored toward the individual needs of each active material to overcome the specific challenges and make use of its full potential.

Lithium–sulfur batteries are promising candidates for high energy density batteries. However, the insulating nature of sulfur, volume changes during charging and discharging, as well as the dissolution of polysulfides have represented strong limits to the technology so far. Park et al. tackled these challenges by preparing graphitic carbon–TiO$_2$ composites via a pilot-scale spray-drying process. After etching of embedded iron NPs, mesoporous SPs with vacancies for sulfur infiltration were obtained. The porous graphitic matrix coped well with volume changes and provided high conductivity, while the embedded TiO$_2$ NPs were able to chemically trap polysulfides. The colocalization of these different components within one SP leads to excellent rate capability and cycle performance.

While the previous examples of SPs in electrode active materials were all related to cathode materials, the potential of SPs to improve anode properties is considered to be even greater. This is a consequence of the challenges that arise due to the formation of irreversible passivation layers. Since for most anode materials the lithiation and delithiation take place outside of the electrochemical stability window of the electrolyte, a passivation layer is formed during the first cycles. The formation of this so-called solid electrolyte interphase (SEI) layer consumes parts of electrolyte and active material, whereby a higher surface area of the active material leads to an increased material consumption. Hence, the increased surface area of NPs represents one of the major drawbacks in terms of irreversible passivation for anode materials (Figure 7a).

![Figure 7](https://example.com/image.png)

**Figure 7.** Schematic illustration of a1) SEI formation on MnO$_2$ nanoparticles compared to nano-MnO$_2$@TiO$_2$ supraparticles and a2,a3) corresponding SEM images after 200 cycles. Scheme of b1) pomegranate-like arranged Fe$_3$O$_4$@C nanoparticles and the resulting breaking of the carbon coating due to lithiation, which leads to cracks in the supraparticle on the microscale, displayed by b2,b3) SEM images. Through etching, c1) time-controlled voids in the nanosized yolk–shell structure buffer the expansion, which prevents cracks in the microstructure, displayed by c2,c3) SEM images. a1–a3) and (b1–c3) Reproduced with permissions. Copyright 2018, Elsevier.
Cao et al. approached this challenge by coating SPs consisting of MnO2 NPs with TiO2 NPs that prevent the direct contact of primary anode active material MnO2 with the electrolyte. The SEI layer (Figure 7a1) formed on the surface of these micrometer-sized SPs and grew only slightly in diameter after deep cycling, while single MnO2 NPs possessed a thick passivation layer, which resulted in significant loss of capacity.[117]

A very similar observation was made by Lee et al. for Fe2O3 NPs as anode material, where the geometric confinement of the nanosized building blocks in SPs improved the cycle stability and rate capability.[118] However, even as NP-based material, Fe2O3 suffers from volume changes during lithiation and de-lithiation. Resulting cracks due to mechanical stress in the microstructure expose fresh active material surfaces (Figure 7b), which are consequently passivated leading to more loss of capacity.[109] Han et al. tackled this challenge by fabricating a pomegranate-like SP consisting of yolk–shell Fe2O3@C NPs. The well-defined void in the nanosized yolk–shell structure (Figure 7c) allows a buffering of the volume changes to prevent cracking of the microsphere.[119]

Further approaches to overcome the drawbacks of Fe2O3 NPs through adjusted SP designs include the embedment into a graphene matrix,[120] as well as the work of Xiong et al., who prepared hollow composite microspheres consisting of Fe2O3 NPs and a crossing net of carbon nanotubes that stabilized these SPs during volume changes.[121]

While most commercial lithium-ion batteries still rely on graphite as anode material, the next generation is proposed to be a composite of graphite and silicon NPs due to the outstanding specific capacity of ≈ 3580 mAh g−1 for silicon, which is about ten times higher than graphite.[122] However, most of the previously discussed challenges for anode materials like volume changes and irreversible passivation are present for silicon as well, and even on a larger scale. In case of full lithiation, silicon undergoes a change in volume of up to 300%.[110,112] The reoccurring cracks cause increasing passivation and consumption of the active material, which is why a short cycle life is the main issue for silicon. A large number of different SP designs have been reported to overcome these major drawbacks of silicon. Most common are various forms of hierarchical composites containing carbon-coated silicon NPs and carbon nanorods,[123] often relying on spray-drying as the processing method of choice.[116,124] To provide an easy conception of the 3D structure, these SPs are sometimes described as raspberry-like,[125] pomegranate-like,[126] or watermelon-inspired.[127] One major advantage of silicon is its extensive abundance. In light of sustainability, the work of Zhang et al. stands out due to its unique way of producing silicon NPs out of rice husk, a common high output agricultural waste.[128] SPs containing these low-cost silicon NPs as well as carbon nanotubes showed remarkable electrochemical performance.

SPs have also been reported for more exotic anode active materials like micrometer-sized assemblies of ZnS/CoS/CoS2@N-doped NPs.[129] spray-dried mesoporous mixed Cu-Ni-oxide@graphene nanocomposites,[130] and nanoporous TiNb2O7/C composite microspheres.[131] Closely related to the lithium-ion technology are sodium-ion batteries that generally possess lower energy densities. However, the availability of sodium is much greater than that of lithium, where natural reservoirs are limited. Therefore, the sodium ion technology is expected to play a crucial role for sustainable energy storage systems in the future.[132,133] Due to similar physicochemical characteristics of sodium and lithium, most of the main challenges in sodium ion batteries are analogous to the herein described issues with lithium-ion batteries, like low conductivities of active materials, as well as volume expansion and the resulting increasing SEI formation on anodes. Hence, the potential that SPs offer to improve this type of battery is comparable to the lithium ion technology. With regard to cathode materials, Shan et al., for instance, produced SPs consisting of Na0.5MnO2 nanorods, carbon nanotubes, and reduced graphene oxide via spray-drying. Via this SP approach, the system exhibited significantly improved conductivities.[134] Similar beneficial aspects, leading to enhanced cycle life and rate performance, could be observed for graphene-scaffolded Na2V3(PO4)3 SPs.[135] In order to cope with volume changes at the anode, SPs that are described as hollow microspheres were reported as promising candidates. For example, Choi et al. prepared hollow SeFe2/graphitic carbon spheres with interconnected multicavitities,[136] whereas Xie et al. synthesized hollow SPs consisting of carbon coated Na2Ti3O7 rods.[137] Alternatively, Kim et al. reported metal sulfide/carbon composite SPs that were synthesized via spray-pyrolysis with NaCl NPs as templates to obtain cubic nanocavities in the final SPs. In case of MoS2/C these particles showed good rate performance and delivered high reversible capacity.[138]

As mentioned at the beginning of this section, next to batteries, also supercapacitors and fuel cells play an important role regarding energy storage devices. Supercapacitors have a key function whenever the required power densities exceed the capabilities of batteries. Compared to batteries, SPs have only been reported on few occasions in this field of research. However, these cases are good examples of how SPs can improve certain characteristics, like energy density[139] or conductivities of active materials.[139] For instance, Yan et al. prepared double-layered yolk–shell SPs where NiCo2S4–Ni9S8 NPs are embedded into an amorphous carbon matrix[140] and Jeyaranjan et al. reported ternary scalable hierarchical microspheres composed of polyaniline nanofibers, reduced graphene oxide and cerium oxide nanorods with significantly improved reversibility of the pseudocapacitive characteristics leading to a greatly extended cycle life.[141]

With regard to fuel cells, SPs can play a key role in the fields of hydrogen production and oxygen evolution reaction.[142] Similar to supercapacitors, SPs in fuel cells have not been part of extensive research yet, but several examples can be found that underlie the future potential of SPs for the technology.[143,144] The improved catalytic properties of these SPs, that are decisive for their application in fuel cells, will be covered in the following section.

2.2.2. Catalysis

In order to use energy in a sustainable way, it is necessary to develop more efficient approaches to convert and to store energy, but also to reduce the consumption of energy for certain chemical/physical processes. The efficiency of such a process can be seen as a measure for its sustainability, taking factors
like reaction rate, required amount of energy and chemicals, selectivity, and yield into account. All these factors are affected by materials that exhibit catalytic activity. The catalysis process itself describes the acceleration of a chemical/physical reaction by reducing the energy barrier the educts must overcome to form a product in a lower energy state.[142]

Nowadays, the chemical industry, as one of the biggest energy-consuming branches, takes our society above “basic human needs” to a higher level of comfort. Thus, the use of catalysts in industrial chemistry is stringently required heading toward a more sustainable society. Furthermore, most industrial established reactions rely on the use of catalysts to be economically feasible.[146] In the last years of the 20th century Anastas et al. already introduced the term “green chemistry” summing up 12 requirements for sustainable energy development in this industrial field.[147] The most relevant requirements regarding catalysis are energy efficiency, employment of less toxic chemicals, waste prevention and the use of renewable raw materials (in this case catalysts).[148] More than two decades later, the urge to shift the development toward sustainability became even larger due to the aggravating global climate crisis. This underlines the research efforts to develop new efficient catalytic materials.

Today, around 80% of economically important chemical reactions involve heterogeneous catalysis.[149] In this case, the catalyst itself acts in another phase than the reactants of the catalyzed reaction, e.g., liquid reactants with solid catalyst. In contrast to homogeneous catalysis, the heterogeneous approach brings along advantages like simple removal of the catalyst, efficient recovery, as well as high control over the material structure and properties itself.[150] One field of intense research effort in heterogeneous catalysis are NPs. Due to the possibility to precisely control size, as well as structure achieving a high surface area and due to the broad range of available materials, NPs exhibit excellent potential in catalytic applications. Still, there are several problems related to their use. The removal of NPs from reactions is usually rather complicated and they tend to agglomerate due to their high surface energy, which deteriorates their efficiency. Furthermore, they exhibit size-dependent physical, biological, and environmental dangers.[151] The assembly of individual NPs into SPs targets to overcome most of these disadvantages. Further, by confining the particles to distinct entities, new properties can arise due to their coupling.[152] One should mention in this context that NP deposition on a micrometer-sized support, which has already been used in catalysis for decades, cannot be considered as SP fabrication as usually no cooperative interplay between two components and no organized structure are achieved.[149] The phenomenon emergence is also crucial for the application of SPs in catalysis as it describes their inner porosity yielding high surface areas and controlled particle morphologies. Dependent of the resulting SP structure (dense, hollow, hierarchical porous, composite nanostructured) the performance of the catalyst is thus directly influenced.[149] However, this has been recently excellently reviewed by Gradon et al.[149] focusing on how to precisely control the structure and morphology of SPs in aerosol-based assembly methods aiming for new catalytic materials and will thus not be discussed in detail herein. Another recently released review article by Hou et al. outlined different methods to create SPs for various catalytic fields.[153] This is why in the following, we only outline a few examples of systems that are of particular importance in light of sustainability.

As already noted, energy conversion systems particularly rely on the efficiency of involved chemical or physical reactions. In fuel cell applications, reactions like hydrogen evolution, oxygen evolution reaction, and oxygen reduction reaction play a crucial role.[142] Electrocatalytically active SPs can greatly affect those reactions. Ro et al. designed efficient electrocatalytically active SPs made of bimetallic Pt/Ni NPs loaded on Fe3O4@SiO2.[143] The SPs featured efficient H2 production up to over 96% of the theoretically estimated value by converting NaBH4 into hydrogen. Their catalytic activity was comparable to those of the single NPs. Using bimetallic Pt/Ni NPs as catalytic species in the SPs reduced the amount of needed noble metal while maintaining a sufficient catalytic activity (Figure 8a). In sight of their magnetic properties, the SPs could be easily recycled by exploiting the colocalization properties of the single entities. Thus, multiple cycles were performed to test the stability and recyclability. The SPs showed less than 25% H2 conversion after 5 cycles due to aggregation of the surface NPs. Therefore, an additional TiO2 shell was added for the protection of the loaded Pt/Ni NPs leading to an enhanced stability with a H2 conversion of 94% after 5 cycles.[143] In proton exchange membrane fuel cells electrocatalysts with high efficiency are needed. Porous Pt/C SPs with a 3D pore network and high Pt loading were fabricated to enhance the catalytic activity. The electrochemical characterization showed that the high surface area and the Pt distribution inside the network increased the electrocatalytic performance of the SPs beyond those of the single materials.[144]

Another elegant strategy to produce a sustainable energy source is photocatalytic hydrogen evolution (PHE).[154,155] Recently, Li et al. developed CoFe2O4@ZnIn2S4 SPs that exhibit efficient PHE up to 800 µmol h−1 g−1.[154] This was achieved by creating a p-n junction on the individual particles due to the combination of p-type CoFe2O4 NPs and n-type ZnIn2S4 microparticles. The PHE performance of these SPs was superior in comparison to the single particles. Because of the increased light absorption and the effective charge carrier separation at the p-n junction (due to coupling effects within the SPs), the particles were also used as efficient photodegradation agents for decomposing organic volatile compounds in water. Furthermore, the CoFe2O4@ZnIn2S4 SPs exhibited excellent photostability and recyclability up to 5 cycles for PHE and photodegradation.[154]

As mentioned above, photocatalytic reactions can not only support energy storage and conversion but also greatly influence water purification by decomposing pollutants, i.e., dyes, volatiles or toxic chemicals. Herein, the catalytic reaction is based on the absorption and use of sunlight’s energy, which is one of the main sustainable energy sources for humankind. Many materials are already known, that exhibit photocatalytic properties, i.e., ZnO,[156] NiO,[156] carbon-based materials,[157] Fe3O4,[158] or Bi2MoO6,[159] and BiVO4.[160] A well-established photocatalyst among these is titanium dioxide (TiO2).[136,161,162,163] Due to its wide bandgap, it is well suited for the absorption of solar light. The active surface area of catalytic materials is an important
factor influencing its reactivity and efficiency. In order to increase the available catalytic surface, TiO$_2$ NPs in combination with polystyrene NPs were used as precursor materials for highly porous TiO$_2$ SPs. Liu et al. synthesized porous TiO$_2$ SPs by a surface-mediated fabrication method and subsequent removal of polystyrene and used these structures as catalysts for photodegradation of rhodamine B.$^{[164]}$ The precise control of pore size and distribution throughout the SPs enables a photocatalytic activity comparable to single NPs while allowing easy removal from the treated solution.$^{[162]}$ This improvement in catalytic behavior can be ascribed to the emergence property of the synthesized SPs. A similar approach was conducted with WO$_3$ as photocatalyst (Figure 8b).$^{[165,166]}$ By adding Pt NPs to the porous WO$_3$ SPs a photodegradation profile superior to the one of single WO$_3$ NPs was achieved.$^{[166]}$ In another approach, magnetic NPs were added to the photocatalytic SPs. Herein,
Fe$_2$O$_4$ was used as a core coated with SiO$_2$ and afterward with TiO$_2$ NPs. By implementing the SiO$_2$ shell around the Fe$_2$O$_4$ core photodissolution of magnetite by TiO$_2$ was prevented. The final SPs exhibited photodegradation of methylene red in water while easy magnetic removal and reuse were possible. [167] The combination of Fe$_2$O$_4$ and TiO$_2$ NPs was also investigated by Shi et al. [163] In an autoclave synthesis they combined these materials forming a supraparticulate structure. By precise control of the reaction temperature, the absorbance ability of the SPs in the visible solar spectrum was optimized. Next to excellent reusability and improved degradation efficiency compared to commercial TiO$_2$ NPs, the photocatalytic performance of the SPs is enhanced by applying an external magnetic field. This leads to the directional arrangement of magnetic particles, resulting in improved solar absorption of the SPs. [163]

The catalytically supported synthesis of industrially important chemicals by SPs (suiting our definition) is a not yet intensively studied field. We believe that there is a high potential for SPs to advance into this field of catalytic application. In the following, we outline research topics, which partly involve SPs as catalysts or could also be addressed by this approach. Catalysis supported synthesis of organic chemicals can promote reactions like the hydrogenation of organic compounds, aldol reactions, or epoxidation. [168–170]

The hydrogenation of CO$_2$ to methanol or olefins is a recently intensively studied topic as it faces the challenge to effectively lower the carbon amount in the atmosphere. Yet, it remains a challenge to utilize CO$_2$ because of its low reactivity. [168] Fe$_2$O$_4$ microspheres were used as catalysts and modified with different Mn contents for CO$_2$ conversion. The designed SPs exhibited excellent hydrogenation of CO$_2$ toward light olefins. With this approach, a yield and selectivity of 19% and 46%, respectively, to light olefins and an overall CO$_2$ conversion of 45% was achieved by modifying the iron oxide with 9% Mn. Herein, manganese acted as a promoter of the CO$_2$ adsorption and the activation of the C=O bonding while the Fe-based material initiated the reduction of CO$_2$ to CO. [172] The close proximity of these materials resulted in a coupling effect of the electronical properties, thus leading to sufficient reaction. In another approach porous Sn/TiO$_2$/graphite carbon microparticles were used for the photocatalytically stimulated reaction of CO$_2$ and water to CO and CH$_4$. The efficient absorption of light and the good catalytic performance was assigned to the unique composition and structural characteristics of the material. [172]

Au@Fe$_2$O$_4$ SPs with a porous structure were designed to efficiently catalyze epoxidation of styrene. The loading of Au NPs onto Fe$_2$O$_4$ particles led to a further enhanced selective oxidation of styrene with a maximum conversion of up to 95%. [170] Furthermore, porous Pd@Ni$_x$Si$_y$O$_z$(OH)$_2$ SPs were synthesized by Zhang et al. for advanced styrene hydrogenation [173] and Yang et al. utilized porous Ti/Zr microspheres with reasonable cycle stability for the efficient synthesis of y-valerolactone, an important platform molecule and biofuel additive, from biobased ethyl levulinate. [174] Niu et al. used Fe$_3$O$_4$@Fe(OH)$_3$ SPs as efficient catalysts for the reaction between different aldehydes and ketones at room temperature. It was shown that the OH-groups of the Fe(OH)$_3$ shell showed good catalytic activity in aldol reactions, while the magnetic Fe$_3$O$_4$ core enabled easy removal and reusability. [169]

All the above-mentioned work can contribute significantly to a more sustainable development of our modern society. The catalytically supported reactions, i.e., the conversion of CO$_2$ or the purification of water by photodegradation, are just the obvious contributions. By creating SPs, properties related to sustainability develop even further, like magnetically removable and reusuable catalysts, porous systems for less material in use or by substituting rare and cost-intensive materials (noble metal catalysts). The unsophisticated removal and the reuse of these SPs make them suitable for a sustainable resource management. Besides, the greatest advantages are the simultaneous retention and the improvement in their catalytic behavior, which arises from their structural and morphological uniqueness.

### 2.3. Material-Based Modern Life Comfort

The use of energy in the form of electricity for the operation of our countless commodities has become an integral part of everyday life in our society. While resource efficiency with respect to “energy” is already part of heavy research efforts, little has been put into systematically investigating material wear and disposal.

Mass production and increasing purchasing power in the course of industrialization led to the formation of a consumer society whose consumption behavior is characterized by a wasteful “take-make-dispose” pattern, i.e., in which durable goods are treated like consumable goods, at cost of environmental pollution and destruction. [175] Ubiquitous seduction marketing and even cultural norms trigger us to buy more and more products that we would not necessarily need to fulfill our basic needs. Once a product breaks down, it is either financially unviable to be repaired or it is simply not designed to be fixable. Consequently, large amounts of goods end up as useless waste in landfills, which results in an enormous loss of valuable material. Therefore, this state-of-the-art linear economic model, which is designed for throughput rather than reusability, cannot be defined as sustainable, since it only functions based on the assumption that resources are permanently available. This does not correspond to reality and therefore does not contribute to the ability to exist constantly.

A novel alternative strategy to address this issue is the concept of circular economy, a system in which ideally waste does not exist. Products are consequently designed to last and are optimized for circulating in closed loops. These loops such as reuse, repair, remanufacture, refurbishment or recycling extend the product’s life cycle and improve resource productivity. This production approach is not only sustainable but also paves the way to resilient economic growth and a systemic answer to reducing dependencies on resource markets due to net material savings, less price volatility, and supply risk. [176]

### 2.3.1. Extending Materials Lifetime

In order to avoid waste in the first place, it would be advantageous to design products to last. Due to their characteristic coupling, emergence, and colocalization effects, SPs can contribute
to the development of long lasting products, for example, within coatings or as an alternative to pigments.

“Anti-X” Coatings: Most modern products use coatings on their surfaces to protect the materials they are made of and thus, enhance their durability. While antiscratch, antiwear or anticorrosion coatings prevent damage of the coated material, antifouling, anti-soiling, self-cleaning, and antifogging coatings mainly avoid a regular cleaning of the material with its potentially negative effects of mechanical and chemical stress on their lifetime.

Nowadays, NPs are already regularly used for coating products. They can provide the same intrinsic properties as their bulk counterparts, such as UV absorption by ZnO or the high refractive index in case of TiO2, while showing less light scattering due to their size, which enables the creation of transparent coatings. However, for the development of coatings where surface wetting plays a dominant role, i.e., for coatings that should provide an easy removal—or prevent the accumulation—of substances like oil, water, or dirt on them, the use of NPs often does not result in the desired surface structure of the coating. This is when SPs provide a promising alternative due to their larger size dimensions. Coatings that require this feature are for example anti-corrosion coatings avoiding water accumulation, self-cleaning coatings enabling easy dirt, water, or oil removal, antisoiling coatings preventing dirt deposition, or antifogging coatings tackling fogging by water vapor. The minimization of surface wetting by diverse matters can be realized via superhydrophobic or superamphiphobic (e.g., for self-cleaning applications) but also via superhydrophilic (e.g., for antifogging applications) surfaces.

Raspberry-like SPs consisting of one larger surrounded by many smaller spherical particles have gained increasing attention in the coating community due to their unique structure. Coatings based on raspberry-like SPs provide a dual-size roughness due to a coarse structure of about 10 to 20 µm and a finer structure on top of it of about 0.1 to 1 µm. This structure, mimicking a lotus leaf surface, increases the liquid-repellency of a material. Furthermore, the raspberry-like particle structure provides many re-entrant surfaces, which are surfaces hidden under overhangs, on the bottom half of each SP. These enable a vertical stabilization of droplets hindering their sagging into cracks and air pockets of the rough coating structure. Both mentioned characteristic functional properties of these SPs are counted among the phenomenon of emergence.

As examples for SP-based coatings, raspberry-like silica-poly(divinylbenzene-co-glycidylmethacrylate) SPs were developed for superhydrophobic coatings, poly(ethyl methacrylate)-silica ones for antifouling coatings, hollow silica (Figure 9a), and polystyrene-silica ones for antifogging coatings, as well as polystyrene ones for superamphiphobic coatings (Figure 9c).

While dual-size hierarchical structures could also be produced via layer by layer assembly, templating methods, chemical vapor deposition, lithography or plasma etching, the use of SPs is of advantage as no specialized equipment is needed for the coating procedure. In comparison to the application of simple spherical (micro)particles, which would also yield a coating with rough surface structure, raspberry-like SPs provide
Photonic Colors: In contrast to other functional surface coatings, paints/pigments mostly meet rather aesthetic purposes than a certain surface property. Furthermore, paints and their use are deeply connected to mankind starting from cave paintings in the Stone Age.\(^{[187]}\)

This is why colorants are ubiquitous in modern society and everyday life as they are used for many applications like food, clothing, cosmetics or paints.\(^{[188]}\) The controlled tuning of hue and coloring effects, which is essential to fulfill various aesthetic needs, is achieved by the regulation of scattering and absorption properties of the colorant.\(^{[189]}\) For manmade colored products, absorption is usually manipulated using organic dyes and pigments. Although they offer a broad variety of colors, there are several drawbacks, such as photo- or chemically-bleaching.\(^{[190,191]}\) Additionally, in order to achieve different hues and optical effects, molecular dyes often have to be combined with supplementary compounds. A broad range of colors hence results in a multitude of chemical compounds, which increases costs of production and requires higher numbers of toxicity tests when brought into contact with the human body, for instance.\(^{[189]}\)

Besides the coloration of fabricated products, astonishing examples and a large variety of colors are found in nature.\(^{[192]}\) All living organisms, such as animals and plants, as well as several minerals show coloration effects, often caused by their structure.\(^{[187,193,194]}\) These structurally colored materials, also called photonic pigments or photonic balls in the case of colloidal NPs that serve as building blocks with sizes in the scale of the wavelength of visible light.\(^{[187,195]}\) Thus, scattering appears due to Bragg’s law, i.e., a certain wavelength shows angle-dependent, constructive interference depending on the primary particle size.\(^{[12,196,199]}\) This effect appears macroscopically as coloration to the human eye. The wavelength of the constructive interference and hence of the coloration is shiftable over the entire visible spectrum by adjusting the size of the colloidal building blocks, which are often composed of silica or polymers.\(^{[197]}\) In contrast to organic dyes or inorganic pigments, which result in coloration effects due to absorption and, thus, rely on the materials chemistry, structural coloration can be generated based on a single inert material.\(^{[190]}\)

Incoherent light scattering significantly reduces color purity and brightness. Moreover, the combination of structural elements on the scale of visible light with light-absorbing molecular features has been shown to increase the brightness and clear hue. An important method to overcome the undesired influence of incoherent scattering events is to add a wide-band absorbing component, such as carbon black or iron oxide to the colloidal system.\(^{[193]}\) The black absorbing species absorbs light over the entire visible region due to its intrinsic material properties, thus purifying and increasing the color saturation of the photonic pigment. Therefore, vivid colored pigments can be tailored by the combination of NPs with controlled size and a black absorbing species. In addition to the absorption of undesired incoherently scattered light, these second species of building blocks can be used to tune the inter-scattering distance, i.e., the distance between silica NPs within the SPs.

Recently, Jiang et al. published their work on the addition of iron oxide NPs as the second particle species, which serve as a black absorber and spacer between the scattering silica NPs (Figure 10a). They were able to adjust the SPs color by controlling the size of the silica NPs and the addition of iron oxide NPs permitted a better manipulation of the resulting hue and brightness of the tailored color (Figure 10aL1,a2).\(^{[194]}\) The absorbing species does not have to be black necessarily; it needs to absorb incoherently scattered light over the complete optical spectrum. Therefore, besides iron oxide NPs, silver NPs are well suited as additive for color purification as well. Lai et al. succeeded in the synthesis of different colored photonic pigments (Figure 10b1,b2) by the assembly of different sized polystyrene NPs (Figure 10b3), which is another striking example of the emergent property of structural coloration in SPs assembled from smaller, nanoscale building-blocks. The hue and the brightness of these were significantly increased by the addition of silver NPs (Figure 10b1).\(^{[200]}\)

While photonic SPs can be generated via colloidal assembly in a slowly evaporating droplet,\(^{[12,194,201]}\) microfluidic fabrication is also suitable for the creation of structural colors.\(^{[202]}\) Park et al. have shown that microfluidic techniques can generate microcapsules containing densely packed particles, such as polystyrene/poly(N-isopropylacrylamide-co-acrylic-acid) core–shell particles (Figure 10c1,c3). The core size and hence the size of scatterers, as well as the shell diameter can be well controlled, which enables the precise adjustment of the wavelength of coherently scattered light and thus, the coloration of the microcapsules (Figure 10c1,c2).\(^{[190]}\)

Comparing photonic pigments to conventional chemically colored pigments, photonic pigments exhibit some major advantages, especially in terms of (mechanical, chemical, thermal, ...) stability and thus in terms of sustainability.\(^{[191,203]}\) Since the coloring effects are generated from scattering events with nanostructured features and the nanoscale building blocks often consist of chemical- and photo-stable materials, photonic pigments usually offer nonfading properties. Therefore, they keep their intense color and brightness over time.\(^{[196]}\) On the contrary, chemicals of conventional colorants eventually degrade and this can cause environmental pollution and can add additional future work; as well as costs for repainting.\(^{[191,199]}\) Therefore, the lifetime of products painted with photonic pigments as colorant can be increased dramatically. Furthermore, in contrast to conventionally used pigments, photonic pigments are not composed of any heavy metals or carcinogenic substances.\(^{[195,204]}\) Finally yet importantly, photonic pigments made from biological, environmentally benign, nanoparticulate building blocks offer a great, futuristic alternative to conquer
the rising demands for sustainable, stable, and nontoxic colorants.\cite{187,193,199,203}

2.3.2. Bringing Materials Back in Use

While the extension of materials lifetime certainly should be the goal for all products in use, wear, and tear or damage, caused by environmental influences, cannot be completely prevented.

Hence, easy maintenance to keep a product in working condition via simple detection and replacement of broken components must be implemented to avoid functional obsolescence and subsequently material dissipation. Once a product cannot be repaired anymore, at least the recycling and reuse of its material components is desired to save limited resources on our planet.

Sensors for Easy Repair: The use of sensor or indicator materials reporting physical or chemical damage due to, e.g., mechanical stress, temperature, aggressive solvents, pH changes or chemical gases is of increasing importance for the sustainable use of products as it enables defect indication of sub-micrometer sized components and, thus, their targeted repair.

For the development of sensor materials for harmful environmental triggers, NPs are already known to offer various outstanding properties in this regard compared to bulk materials. Besides their advantages already outlined in this review before (Section 2.1.1.), the possibility to miniaturize sensors\cite{28} enables their integration into smallest product components and into almost any type of material, which is crucial for the development of sustainable sensor-equipped products.

As sensors are desired to detect several different triggers, SPs exhibit a variable platform to bottom-up design the desired properties and thereby possess functionalities that go beyond the single NP properties. This has been explained in detail for gas sensing applications (Section 2.1.1) but also holds for every type of sensor leading to high sensitivity and selectivity of these...
SPs. A few research groups\textsuperscript{205–207} even demonstrated that SP-based sensors can provide a response to analyte binding or mechanical forces through structural reorganization or fragmentation of their components which is an advanced sensor behavior in comparison to nanomaterial-based sensors that normally show a direct change of their physical properties.

In this context, Osterloh et al. prepared silica–gold SPs via electrostatic self-assembly as sensors for various linear chain alkanethiols.\textsuperscript{205} They show a color change in presence of the analyte due to reorganization of the gold NPs on the silica core particle (Figure 11a). Examples of gas detecting SPs have already been described in Section 2.1.1. extensively and may be easily transferred to sensor applications for easy product maintenance. An SP-based pH sensor has been created through the embedding of magnetic NPs and indicator dyes within a polymer particle by a miniemulsion solvent evaporation technique.\textsuperscript{28} A temperature-sensitive SP was shown by Lee et al. based on CdTe NPs connected to a gold NP by poly(ethylene glycol) derivatives.\textsuperscript{206} It monitors temperature changes between 20 and 60 °C via variation of the luminescence intensity (Figure 11b). Cold chain breach indicating magnetic supraparticulate microrods have been described by Müssig

![Figure 11. SEM images of silica–gold supraparticles as sensors for alkanethiols in a1) the as-prepared state and a2) after contact to propanethiol showing a3) a color change in the visible wavelength range dependent of the analyte type. Scheme of b1) Au–poly(ethylene glycol)–CdTe supraparticles, as well as b2) TEM. b3) This sensor indicates temperature changes via luminescence intensity variations. Scheme and SEM images of dye-doped silica–iron oxide supraparticles as shear stress indicators in c1) the as-prepared state and c2) after abrasion reporting c3) the damage via an increase in fluorescence intensity. (a1–a3) Reproduced with permission.\textsuperscript{205} Copyright 2004, American Chemical Society. (b1,b2 and c1–c3) Reproduced with permission.\textsuperscript{206,207} Copyright 2005 and 2020, Wiley-VCH GmbH & Co. KGaA.](image-url)
et al.[208] Besides other mechanical stress indicator SPs,[209] a shear indicator based on hierarchically structured luminescent dye-doped silica NPs and iron oxide NPs assembled via spray-drying has been established.[207] It reports damages, e.g., by an increase in fluorescence signal (Figure 11c).

In summary, sensor and reporter SPs may play a key role in the development of easy maintenance products in the future due to their high sensitivity and selectivity based on the phenomena of emergence and colocalization offering high porosity as well as the combination of diverse materials within a sensor. However, also sustainable, repair-friendly product design needs to become the standard to make these SPs a powerful tool.

**Tracer-Based Sorting as Recycling Aid:** Once materials are not working properly anymore and cannot be repaired, their recycling and reuse are desired to save limited resources on our planet and thereby act sustainably. A major challenge for recycling is the automated sorting of different materials, such as polymers, to receive a high purity recyclate.[210] If polymers are not sorted properly, the resulting recyclate exhibits insufficient material properties and limits its reusability.[211] Thus, the efficient sorting of different materials is a crucial step for the recycling of waste streams.

The direct sorting utilizes intrinsic properties of the materials such as density, near-infrared fingerprint or magnetism but is often unsuitable for automated recycling especially when composite materials are to be recycled.[212] An alternative is the addition of a tracer into materials.[211] This tracer-based sorting often utilizes microparticles which are incorporated into plastics during their production and enables the identification of plastic by distinct (most frequently optical) signals during sorting and recycling.[213]

To cope with the mixed nature of the waste and thereby increase the recovery rates, tracers with more than one detectable signal are desired.[212] In contrast to NPs or microparticles, SPs represent a flexible platform to create multifunctional tracers. Furthermore, they permit signal variations[214] and thereby to distinguish and sort different materials. Several reviews focus on the idea of object identification with nano- or microscale tracers in general.[215] In terms of SPs for recycling, only one publication from our group was found to date where SPs were described to contribute to recycling, indicating high capabilities for future contributions in this field.[216] Therein, a magnetic SP is suggested to enable sorting and identification of dark plastics. By variation of the composition, different magnetic fingerprints are resolved via magnetic particle spectroscopy (Figure 12a). Such a magnetic signal is advantageous for colorful and dark materials because it is not inferred by light absorption in the visible range and can thus be detected from within materials. Although not explicitly mentioned in many manuscripts, other SPs exhibit potential for their utilization in recycling as well.

One principle of signal variation is a structural variation within SPs. These codes are often referred to as graphical codes. It is important to note that the graphical signal is resolved within one SP rather than the arrangement of multiple SPs. Because materials in waste streams are often shredded the code generated by the spatial arrangement of microparticles may thereby be destroyed. A defined pattern of different materials is used to generate a barcode information, which is resolved, for instance, due to different reflectance, sizes or magnetic properties. Progress in this field is well reported in other articles and is thus not further discussed herein.[217]

A SP that combines such a graphical patterning with a spectral signal was shown by Lee et al. Herein, the use of rare-earth upconverting nanocrystals reduces spectral interference from the sample due to large anti-Stokes shifts.[218] Such upconverting luminescent spectral signatures can be tailored by variations of lanthanide composition, dopant concentration, mass content,[218] or by addition of dyes into the SP (Figure 12b),[220] yielding a fluorescent resonant energy transfer due to coupling. SPs based on encoded silica beads[221] (Figure 12c) or polymers[222] with quantum dots are another class of luminescent signal carriers. The different colors of various quantum dots are resolved via fluorescence imaging or fluorescence spectroscopy. Gao et al. suggested to detect the optical properties of quantum dot encoded polystyrene beads with flow cytometry, yielding a rapid readout. However, this system was only demonstrated in liquids and might be limiting for recycling applications.[223]

Song et al.[224] utilized the flexibility of SPs by layer-by-layer assembly of quantum dot NPs around gold NP cores. Thereby, both the intrinsic optical and plasmonic properties of quantum dots and gold NPs are preserved and enable the detection of both signals due to colocalization within the SP.

As an alternative to layer-by-layer assembly or swelling techniques, microfluidic syntheses of quantum dot encoded SPs were reported[225,226] as a simple and cost-effective method.[225] The microfluidic synthesis of barcode particles in general is reviewed in another excellent article.[227]

As SPs provide the possibility for variation of its NP building blocks, it facilitates the incorporation of quantum dots into microbeads in defined ratios. The resulting signal represents the ratio of incorporated quantum dots due to their colocalization and is thus controlled by the structure of the SP.[228] A similar principle was used to create luminescent IDs based on different rare-earth doped CaF2 NPs that were assembled into SPs in defined ratios (Figure 12d).[229]

As an alternative to optical signals, SPs based on surface-enhanced Raman spectroscopy (SERS/SERRS)-encoded NPs[230] or encapsulated DNA[231] were reported. In order to decode the DNA via quantitative real-time PCR and Sanger sequencing, the SPs were equipped with magnetic NPs that enable the magnetic collection of the tracer. While DNA codes provide a large variety of possible codes and need only a very small amount of tracer, they are to date still expensive to decipher.

Magnetic iron oxide NPs were also combined with quantum dots[221] or upconverting nanocrystals[231] yielding multifunctional SPs. Thereby, the optical fingerprint of the SPs was maintained while adding the possibility of magnetic separation[231] or enabling magnetic resonance imaging.[212] Another work expanded this even further and presented a dual-mode encoded magnetic SP by combination of fluorescence reporters and SERS probes[234] and thereby represents a true multisensor tracer which will be desired for future recycling applications.

These examples show the high potential of SPs for tracer-based sorting as recycling aids and may thus inspire more researchers to focus on their development and implementation into recyclable materials.
All in all, the use of SPs in products may not only enhance their durability, resulting in fewer product damages and a prolonged product lifetime, but also enable their easy maintenance as well as the recycling of their components. This can ultimately help us to bring the abstract concept of circular economy to life.

3. Conclusion

Sustaining existence of humans on our planet Earth in the future demands serious efforts in many fields and dimensions. Technology is a crucial and in fact key contributor to secure the fulfillment of human needs in line with ensuring the ability for us to exist constantly, i.e., to achieve a sustainable living on the planet. As outlined, for any technology, advanced materials are decisive contributors on all levels to secure “fundamental needs,” as well as to enable “basic” and even “modern life comfort” in the future as (at least the industrialized) nations are taking it for granted today. Focus in this review was put on how the merits of NPs to contribute to sustainability can be enhanced further by combining them to entities called SPs. By doing so, three fundamental new properties, namely, coupling, emergence, and colocalization occur and yield new functionalities. These contribute to the defined sustainability topics “purification technologies,” “agricultural delivery systems,” “energy storage and conversion,” “catalysis,” “materials with extended lifetime,” and “materials brought back in use.”

In the fields of “energy storage” and “catalysis,” the combination of NPs to SPs to enhance properties or even achieve completely new functionalities has been already and is currently even more heavily in the focus of research and impressive systems were reported.

In the other fields studied in this review, we see great potential for SPs to enhance properties or even achieve completely new functionalities has been already and is currently even more heavily in the focus of research and impressive systems were reported.

In the other fields studied in this review, we see great potential for SPs to enhance properties or even achieve completely new functionalities has been already and is currently even more heavily in the focus of research and impressive systems were reported.

Several general challenges for establishing the use of SPs in these different fields of sustainability that should be particularly
addressed in future are, according to the authors’ perspective, as follows:

- From a scientific perspective, many assembly processes are still not very well understood and the obtained SPs are thus only achieved via empirical studies. Calculations and simulations could help to fully understand SP formation and predict emergent sample properties. This would ultimately enable the tuning of internal or external assembly parameters to engineer SPs in a very reliable and predictable fashion.[233]

- Regarding the transition from chemical lab-scale SP synthesis to industrial fabrication, the main challenges are fabrication time, scalability, and cost. Many assembly processes require large time as NPs have to come together, rearrange and form ordered structures, which are kinetically restricted processes due to small diffusion rates of the building blocks.[236] A high number of SP syntheses are difficult to scale-up due to primary NP yield, matter transport at larger volumes, or the presence of competing processes, which scale up differently.[235] Finally, the added value of the use of SPs compared to NPs or random agglomerates to an application must equal out the surely higher fabrication costs of these materials.[236]

- In order to truly contribute to sustainability, also the fabrication processes of SPs should be as little energy-consuming and the starting, as well as processing materials, as green, abundantly available and recoverable as possible. Furthermore, the reuse and recycling of SPs themselves after their application in a specific field should be critically reviewed.

In general, we see that the increase in SP research during the last years was enormous and we think that SPs as versatile functional systems have just started their triumph. Thus, borrowing the words of Feynman,[237] we think particularly with the last years was enormous and we think that SPs as versatile functional systems have just started their triumph. Thus, borrowing the words of Feynman,[237] we think particularly with the

Acknowledgements

This work was financially supported by the BMBF (NanoMatFutura grant 03XP0149), which is gratefully acknowledged. J.R. and S.M. acknowledge their scholarship funding of the German Federal Environmental Foundation (DBU).

Open access funding enabled and organized by Projekt DEAL.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

nanomaterials, nanoparticles, supraparticles, sustainability

Received: January 7, 2021
Published online: February 1, 2021

[1] P. E. Shaw, L. Newby, Regul. Stud. 1998, 32, 863.
[2] G. H. Brundtland, Environ. Policy Law 1985, 14, 26.
[3] T. Kuhlman, J. Farrington, Sustainability 2010, 2, 3436.
[4] Nanotechnology Toward the Sustainocene (Ed: T. A. Faunce), Pan Stanford Publishing, Singapore 2015.
[5] B. D. Fath, Int. J. Des. Nat. Ecodyn. 2017, 12, 1.
[6] a) T. Pradeep, Anshup, Thin Solid Films 2009, 517, 6441; b) X. Hu, G. Li, J. C. Yu, Langmuir 2010, 26, 3031.
[7] M. Segev-Bar, H. Haick, ACS Nano 2013, 7, 8366.
[8] E. Roduner, Chem. Soc. Rev. 2006, 35, 583.
[9] S. C. Glotzer, M. J. Solomon, Nat. Mater. 2007, 6, 557.
[10] J.-M. Lehn, Chem. Soc. Rev. 2017, 46, 2378.
[11] a) Y. Xia, Z. Tang, Chem. Commun. 2012, 48, 6320; b) T. Wang, D. LaMontagne, J. Lynch, J. Zhiuang, Y. C. Cao, Chem. Soc. Rev. 2013, 42, 2804; c) J. Guo, W. Yang, C. Wang, Adv. Mater. 2013, 25, 5196; d) E. Piccinini, D. Pallarola, F. Battaglini, O. Azzaroni, Mol. Syst. Des. Eng. 2016, 1, 155; e) D. Lawless, S. Kapoor, D. Meisel, J. Phys. Chem. 1995, 99, 10329; f) O. D. Velev, K. Furusawa, K. Nagayama, Langmuir 1996, 12, 2374.
[12] V. Rastogi, S. Melle, O. G. Calderón, A. A. García, M. Marquez, O. D. Velev, Adv. Mater. 2008, 20, 4623.
[13] S. Wintzheimer, T. Granath, M. Oppmann, T. Kister, T. Thai, T. Kraus, N. Vogel, K. Mandel, ACS Nano 2018, 12, 5093.
[14] Z. Xue, P. Wang, A. Peng, T. Wang, Adv. Mater. 2019, 31, 1801441.
[15] a) A. Othman, P. Vargo, S. Andreescu, ACS Appl. Nano Mater. 2019, 2, 7008; b) Y. Zhang, W. Qiu, Y. Ma, Y. Luo, Z. Tian, G. Cui, F. Xie, L. Chen, T. Li, X. Sun, ACS Catal. 2018, 8, 8540.
[16] a) Z. Mao, H. Xu, D. Wang, Adv. Funct. Mater. 2010, 20, 1053; b) Z. Xue, C. Yan, T. Wang, Adv. Mater. 2019, 31, 1807658.
[17] E. W. Chuan Lim, R. Fing, J. Chem. Phys. 2012, 136, 124109.
[18] a) G. Shaddick, M. L. Thomas, P. Mudu, G. Ruggeri, S. Gumy, npj Clim. Atmos. Sci. 2020, 3, 7; b) S. J. Lade, W. Steffen, W. de Vries, S. R. Carpenter, J. F. Donges, D. Gerten, H. Hoff, T. Newbold, K. Richardson, J. Rockström, Nat. Sustain. 2020, 3, 119.
[19] T.-T. Chau, K.-Y. Wang, Sci. Rep. 2020, 10, 2321.
[20] M. S. Mauter, I. Zucker, F. Perreault, J. R. Werber, J.-H. Kim, M. Eilmelech, Nat. Sustainability 2018, 1, 166.
[21] H. Ren, P. Koshy, W.-F. Chen, S. Qi, C. C. Sorrell, J. Hazard. Mater. 2017, 325, 340.
[22] A. Bigdeli, F. Chasemi, H. Golmohammadi, S. Abbasi-Moayed, M. A. F. Nejad, N. F. Kasmani, S. JafariNejad, M. Shahrajabian, M. R. Hormozi-Nezhad, Nanoscale 2017, 9, 16546.
[23] K. Suematsu, Y. Shin, Z. Hua, K. Yoshida, M. Yuasa, T. Kida, K. Shimanoe, ACS Appl. Mater. Interfaces 2016, 6, 5319.
[24] G. T. Chandran, X. Li, A. Ogata, R. M. Penner, Anal. Chem. 2017, 89, 249.
[25] Z. Farka, T. Jufl, D. Kovl, L. Trnkv, P. Skld, Chem. Rev. 2017, 117, 9973.
[26] G. Zhu, C. Xi, H. Xu, D. Zheng, Y. Liu, X. Xu, X. Shen, RSC Adv. 2012, 2, 4236.
[27] Y. Zhang, Q. Xiang, J. Xu, P. Xu, Q. Pan, F. Li, J. Mater. Chem. 2009, 19, 4701.
[28] G. Mistlberger, A. L. Medina-Castillo, S. M. Borisov, T. Mayr, A. Fernández-Gutiérrez, J. F. Fernandez-Sanchez, I. Kliment, Microchim. Acta 2011, 172, 299.
[29] K. Koren, G. Mistlberger, D. Aigner, S. M. Borisov, A. Zankel, P. Pölt, I. Kliment, Monatsh. Chem. 2010, 141, 691.
[30] a) N. Singh, R. K. Gupta, P. S. Lee, ACS Appl. Mater. Interfaces 2011, 3, 2246; b) C. Pannek, T. Vetter, M. Oppmann, C. Weber, A. Eberhardt, M. Dold, M.-L. Baurersfeld, M. Henfling, S. Trupp, B. Schug, J. Wollenstein, K. Mandel, Sens. Actuators, B 2020, 306, 127572; c) S. Bai, K. Zhang, L. Wang, J. Sun, R. Luo, D. Li, A. Chen, J. Mater. Chem. A 2014, 2, 7927.
[31] Y. Shen, W. Wang, X. Chen, B. Zhang, D. Wei, S. Gao, B. Cui, J. Mater. Chem. A 2016, 4, 1345.
[208] S. Müssig, T. Granath, T. Schembri, F. Fidler, D. Haddad, K.-H. Hiller, S. Wintzheimer, K. Mandel, ACS Appl. Nano Mater. 2019, 2, 4698.
[209] a) S. Wintzheimer, S. Müssig, S. Wenderoth, J. Prieschl, T. Granath, F. Fidler, D. Haddad, K. Mandel, ACS Appl. Nano Mater. 2019, 2, 6757; b) S. Wintzheimer, J. Reichstein, S. Wenderoth, S. Hasselmann, M. Oppmann, M. T. Seuffert, K. Müller-Buschbaum, K. Mandel, Adv. Funct. Mater. 2019, 29, 1901193.
[210] S. P. Gundupalli, S. Hait, A. Thakur, Waste Manage 2017, 60, 56.
[211] J. Woidasky, J. Moesslein, P. Wendler, D. Kirchenbauer, D. Wacker, G. Gao, C. Lang-Koetz, Chem. Ing. Tech. 2020, 92, 441.
[212] S. R. Ahmad, Environ. Technol. 2004, 25, 1143.
[213] G. Gao, A. Turshatov, I. A. Howard, D. Busko, R. Joseph, D. Hudry, B. S. Richards, Adv. Sustainable Syst. 2017, 1, 1600033.
[214] a) Y. Xia, Z. Tang, Adv. Funct. Mater. 2012, 22, 2585; b) L. Scarabeli, M. Schumacher, D. Jimenez de Aberasturi, J.-P. Merkl, M. Henriksen-Lacey, T. Milagres de Oliveira, M. Janschel, C. Schmidtke, S. Bals, H. Weller, L. M. Liz-Marzán, Adv. Funct. Mater. 2019, 29, 1809071.
[215] a) S. Shikha, T. Salem, J. Cheng, Y. Zhang, Chem. Soc. Rev. 2017, 46, 7054; b) D. Paunescu, W. J. Stark, R. N. Grass, Powder Technol. 2016, 291, 344.
[216] S. Müssig, F. Fidler, D. Haddad, K.-H. Hiller, S. Wintzheimer, K. Mandel, Adv. Mater. Technol. 2019, 4, 1900300.
[217] a) D.-S. Wang, A. Mukhtar, K.-M. Wu, L. Gu, X. Cao, Materials 2019, 12, 3908; b) B. C. Park, Y. K. Kim, Met. Mater. Int. 2017, 23, 413.
[218] J. Lee, P. W. Bisso, R. L. Srinivas, J. J. Kim, A. J. Swiston, P. S. Doyle, Nat. Mater. 2014, 13, 524.
[219] Q. Zhang, X. Wang, Y. Zhu, J. Mater. Chem. 2011, 21, 12132.
[220] L. Bao, H. You, L. Wang, L. Li, R. Qiao, Y. Zhang, Y. Zhong, Y. Xiong, Z. Li, J. Mater. Chem. C 2014, 2, 8949.
[221] Q. Ma, I. C. Serrano, E. Palomares, Chem. Commun. 2013, 47, 7071.
[222] a) J. Yang, S. R. Dave, X. Cao, J. Am. Chem. Soc. 2008, 130, 5286; b) Y. Zhang, A. R. Clapp, RSC Adv. 2014, 4, 48399.
[223] X. Gao, S. Nie, Anal. Chem. 2004, 76, 2406.
[224] F. Song, P. S. Tang, H. Durst, D. T. Cramb, W. C. W. Chan, Angew. Chem., Int. Ed. 2012, 51, 8773.
[225] X.-H. Ji, N.-G. Zhang, W. Cheng, F. Guo, W. Liu, S.-S. Guo, Z.-K. He, X.-Z. Zhao, J. Mater. Chem. 2011, 21, 13380.
[226] C.-G. Yang, L. Cheng, W.-Q. Ye, D.-H. Zheng, Z.-R. Xu, Colloids Surf., A 2020, 588, 124373.
[227] Y. Zhao, Y. Cheng, L. Shang, J. Wang, Z. Xie, J. Gu, Small 2015, 11, 151.
[228] M. Han, X. Cao, J. Z. Su, S. Nie, Nat. Biotechnol. 2001, 19, 631.
[229] F. Miller, S. Wintzheimer, T. Reuter, P. Groppe, J. Prieschl, M. Retter, K. Mandel, ACS Appl. Nano Mater. 2020, 3, 734.
[230] C. Fernández-López, C. Mateo-Mateo, R. A. Alvarez-Puebla, J. Pérez-Juste, I. Pastoriza-Santos, L. M. Liz-Marzán, Langmuir 2009, 25, 13894.
[231] M. Puddu, D. Paunescu, W. J. Stark, R. N. Grass, ACS Nano 2014, 8, 2677.
[232] Q. Ma, Y. Nakane, Y. Mori, M. Hasegawa, Y. Yoshioka, T. M. Watanabe, K. Gonda, N. Ohuchi, T. Jin, Biomaterials 2012, 33, 8486.
[233] Y. Zhang, C. Dong, L. Su, H. Wang, X. Gong, H. Wang, J. Liu, J. Chang, ACS Appl. Mater. Interfaces 2016, 8, 745.
[234] R. Li, Y. Zhang, J. Tan, J. Wan, J. Guo, C. Wang, ACS Appl. Mater. Interfaces 2016, 8, 9384.
[235] J. K. Stolarczyk, A. Deak, D. F. Brougham, Adv. Mater. 2016, 28, 5400.
[236] O. D. Velev, S. Gupta, Adv. Mater. 2009, 21, 1897.
[237] R. P. Feynman, Eng. Sci. 1960, 23, 22.

Susanne Wintzheimer is postdoc and group manager of the Supraparticle Group at the Friedrich-Alexander University Erlangen-Nürnberg, Germany, since 2020. Since 2021, she additionally is Scientific Head of the Particle Technology Group at the Fraunhofer Institute for Silicate Research ISC in Würzburg, Germany. She holds a Ph.D. in chemistry (graduated: 2017) from the University of Bordeaux, France. Her research interest is especially in the combination of organic and inorganic molecules and inorganic nanoparticles within supraparticles. To date she has published about 18 papers.

Jakob Reichstein studied functional materials at the Julius-Maximilians University of Würzburg, Germany, and received his M.Sc. degree in 2019. Consecutively, he started his Ph.D. with Prof. K. Mandel. Currently, he is a Ph.D. scholarship holder of the German Federal Environmental Foundation (DBU) and research associate in Inorganic Chemistry at the Friedrich-Alexander University Erlangen-Nürnberg. His research focuses on sensor- and indicator supraparticles based on various luminescent nanoparticles for recording harmful environmental triggers.
Philipp Groppe is a Ph.D. candidate in the group of Prof. K. Mandel and research associate in Inorganic Chemistry at the Friedrich-Alexander University Erlangen-Nürnberg, Germany. He studied functional materials at the Julius-Maximilians-University Würzburg, Germany and graduated with a M.Sc. in 2020. His current research focuses on the assembly of porous supraparticles and their beneficial use in various applications, such as in catalysis or as adsorber materials.

Andreas Wolf is a PhD candidate working within a cooperation of the Fraunhofer Institute for Silicate Research ISC in Würzburg and the group of Prof. Karl Mandel at the Friedrich-Alexander University Erlangen-Nürnberg. He has been a research assistant at ISC since 2014 and contributed to different projects in the field of lithium-ion batteries. After further work on various topics at the University of New Mexico, the BMW Group and the Norwegian Institute for Energy Technology he finished his Masters degree in material science at the Julius-Maximilians University Würzburg in 2020. His current focus lies on recycling of lithium-ion batteries.

Bastian Fett is currently doing his Ph.D. in the group of Prof. Karl Mandel at the Fraunhofer Institute for Silicate Research. He graduated with an M.Sc. in the fields of materials science and entrepreneurship from the Technical University Darmstadt, Germany, and the Aalto University, Finland, in 2019. His current research focuses on new materials for perovskite-based tandem solar cell applications by working on both, contact and absorber materials.

Huanhuan Zhou joined the Supraparticle Group as a Ph.D. candidate in October 2020. She obtained her master's degree from National Center for Nanoscience and Technology, University of Chinese Academy of Sciences in 2020. Her research focuses on the design of spray-dried supraparticles.
Rosa Pujales-Paradela is a postdoctoral researcher and research associate in the Professorship for Inorganic Chemistry (Prof. Mandel) at the Friedrich-Alexander University Erlangen-Nürnberg, Germany. She completed her Ph.D. studies in the University of Coruña, Spain, on the design of novel contrast agents for their use in magnetic resonance imaging (MRI) applications in 2019. Her current research focuses on the combination of different materials into porous supraparticle structures for their use in diverse applications, e.g., optical and magnetic markers, adsorbents, and purification.

Franziska Miller is a Ph.D. candidate and research associate in Prof. K. Mandel’s research group at the Friedrich-Alexander University Erlangen-Nürnberg, Germany. She studied Nanoscience at the Eberhard Karls University Tübingen, Germany and received her M.Sc. in 2018. As part of her studies, she contributed to different projects in the field of functional nano- and supraparticles at the Fraunhofer-Institute for Silicate Research ISC in Würzburg, Germany. Her research focus is currently on the synthesis and characterization of optical ID marker supraparticles assembled from nanoparticle building blocks with distinct optical properties.

Stephan Müssig is a Ph.D. scholarship holder of the German Federal Environmental Foundation (DBU) and research associate in Inorganic Chemistry at the Friedrich-Alexander University Erlangen-Nürnberg, Germany. He graduated with an M.Sc. from the Julius-Maximilians University Würzburg, Germany in 2018. His research focus is currently on magnetic nano- and supraparticles on his way to create a magnetic marker technology for various applications.

Sarah Wenderoth studied chemistry at the Saarland University, Germany, and received her M.Sc. in 2018. Since then, she is a Ph.D. candidate at the Julius-Maximilians University Würzburg. Her current research focuses on the field of synthesis and functionalisation of nanoparticles, and their assembly into supraparticles to create smart and communicating surfaces with respect to mechanical shear stress.
Karl Mandel is Professor for Inorganic Chemistry at the Friedrich-Alexander University Erlangen-Nürnberg, Germany, since 2020, and Head of the Particle Technology Group at the Fraunhofer Institute for Silicate Research ISC in Würzburg, Germany, since 2014. He holds a Ph.D. in chemistry (graduated: 2013) from the Julius-Maximilians University Würzburg, Germany. His research interest is in creating supraparticles—mostly via spray-drying—to find new and unexpected properties and employing them as smart objects to contribute to sustainability.