

Figure 1. Graphical representation of hybrid materials: from their chemical roots to the wide variety of final applications. The numbers indicated for each application correspond to the number of reviews found for “hybrid materials” AND the corresponding application (Web of Science). These figures are intended as a mere estimate of the relative abundance of specialized reviews, but they are not mutually exclusive (i.e., a review on “implants” could also be included in “biomed”).

followed to put together the best properties of inorganic quartz and organic polymers in a new type of synthetic rubber for the sake of national security. Indeed, in January 1945, the USA was still at war with Japan, a circumstance that showed clearly on the cover of the January issue of *Popular Science* magazine, featuring a demonized kamikaze pilot about to attack the reader. Inside, among many other war-related technology breakthroughs, there was an article titled “Here is putty with a bounce.” Silly putty was an unexpected byproduct of the race to convert polysiloxanes from useless compounds into useful silicone materials.⁶ But the use of silicones in non-Newtonian fluid toys or in improved gaskets or sealants was just the beginning. The chemistry of polysiloxanes grew and evolved in parallel to their applications, and more and more complex varieties, like polysiloxanes, were added to the family^{7–11} in a growing trend that has not stopped since then.

The path from intercalation chemistry to hybrid materials is another major example of a field with consolidated fundamental knowledge breaking its own boundaries to grow into a new field within materials science. Thus, the use of layered inorganic phases (typically intercalating simple metal cations) as hosts for the intercalation of complex organic cations and polycations marked the starting point for an explosion of a new type of hybrid chemistry.^{12–15} The new knowledge and the new tools developed along the way led to the discovery of early hybrid materials in unexpected ancient materials such as Maya blue or thin Chinese porcelain. Most importantly, this new knowledge and synthetic and analytical tools led to the blooming of hybrid materials rooted in the intercalation chemistry of silicates, oxides, or other chalcogenides, as well as new layered phases.^{2,12,16–18} In addition, they expanded from cationic to anionic intercalates, as well as neutral and solvent and solvated species.

Sol–gel chemistry constitutes another primary root in the early development of hybrid materials. The French school transitioning from “Chimie du Solide” to “Chimie Douce”¹⁹ provided a solid community of bright scientists setting the basis for the synthesis of new metastable and complex hybrid phases, as well as pioneering sustainable, low-temperature routes to ceramics and hybrid materials.

Clément Sanchez played a seminal role in all these fundamental fields and led their consolidation into the broader field of hybrid materials.^{1,2,5,12,20–23} New approaches were added to the discipline, from new types of materials to new synthetic methods, leading to an unprecedented variety of fields in the field. New materials like those formed by the simple ionic integration of polyanion and/or polycation polymers with charged complexes or clusters^{17,24–26} or the whole emerging field of metal–organic frameworks (MOFs) are hybrid by their own definition.^{27–30} There are also new synthetic approaches, like the use of preformed nanobuilding blocks for the construction of sophisticated hierarchically structured materials.^{12,22,29,31–34}

Variety led to the need for classifications to add some order to the profusion of materials. Type I and type II hybrids and organic–inorganic as well as inorganic–organic hybrid materials were proposed, as will be discussed below. However, far more important than these classifications is an understanding of the factors common to all hybrid materials.

Underlying the search for hybrid materials, we will always find the desire to take advantage of the best properties of each of the components, leading in the most favorable cases to synergic properties that go beyond the simple addition of the component’s properties. This *synergy* has been exemplified and reviewed for diverse types of hybrids³⁵ and is one of the factors common to hybrid materials when we look at the final outcome and resulting properties.

But if we consider the chemical genesis of hybrid materials, we will also find very interesting recurring characteristics which are suggestive of general strategies that can be followed for the synthesis of new hybrids. Thus, the concept of self-assembly is present in a wide range of hybrid materials synthesis,^{12,13,36–42} from spontaneous sol–gel processes to the designed assembly of nano building blocks or the crystal growth of metal–organic frameworks. Whether it is revealed by discovery as in sol–gel growth or realized by design as in the case of MOFs, self-assembly is at the heart of the bottom-up growth of hybrid materials. And yet, another large group of hybrid materials are made through bond-engineering, that is, through the purposeful creation of covalent bonds between the dissimilar components to be combined in the hybrid. This approach,

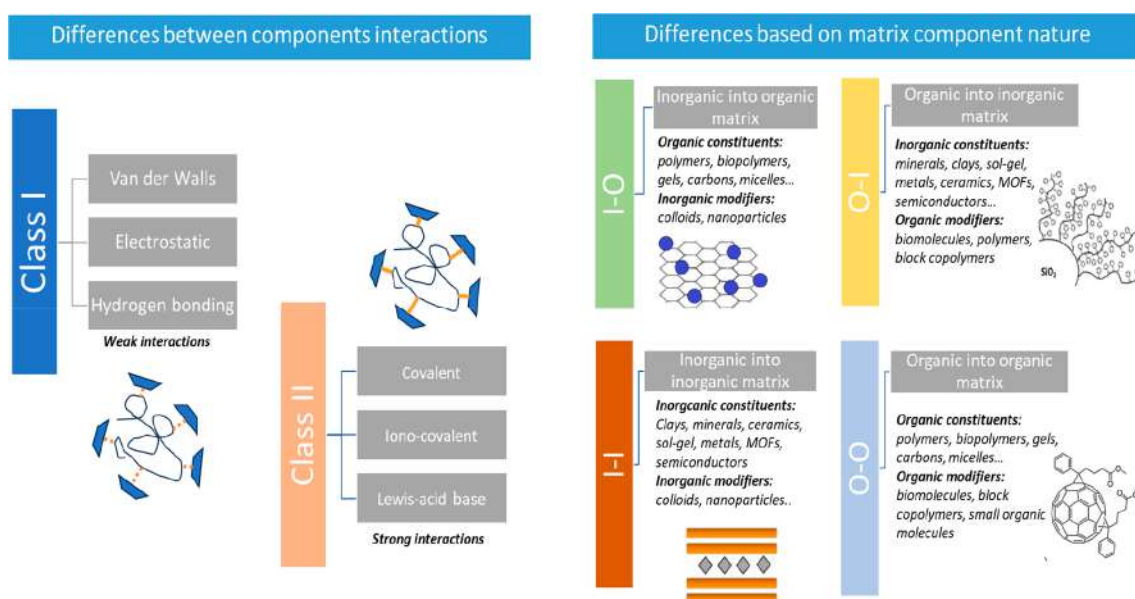


Figure 2. Hybrid materials are general classifications. On the left, classification based on the differences between the interactions of the components, having class I and class II hybrid materials. On the right, the classification according to the matrix and filler component nature, being divided into four classifications: I–O, O–I, I–I, and O–O types.

which has been dubbed “grafting”, represents a radically different approach to spontaneous self-assembly and takes us naturally to the discussion of the wide variety of hybrid materials, their classification, and an attempt to put some Cartesian order in the methods used to synthesize them. Or should we say grow them? Because after this brief introduction, it is already clear that the field of hybrid materials has turned into a complex landscape with a rich orography. The tree of hybrids has turned into a meadow with many trees.

2. HYBRID MATERIALS: A LAND OF MULTIDISCIPLINARITY

“A land of multidisciplinary” was precisely the expression used by Judeinstein and Sanchez in a seminal 1996 paper to describe the emerging field of hybrid materials.²⁰ They appeared as an elegant way to get new materials with multifaceted and tailored features from the combination of organic and inorganic phases at the micro-, meso-, and nanoscale levels.

Two types of classification are commonly used to sort hybrid materials in the literature (Figure 2). The first and most prevalent one is based on the type of interactions between the organic and inorganic components, whereas the second considers which component acts as the dominant matrix and which one acts as guest.

The first classification, originally proposed by Judeinstein and Sanchez,²⁰ considers Class I hybrids as those formed through weak interactions between the organic and inorganic components. This includes hydrogen bonding, electrostatic, and/or van der Waals interactions. On the other hand, class II hybrids are based on strong chemical interactions such as covalent bonds. However, this classification sometimes is ambiguous because, in the same hybrid material, strong and weak interactions can coexist.

Class I hybrid materials are commonly prepared by sol–gel processes, self-assembly, and in situ polymerization methods. In this sense, sol–gel synthesis, developed in 1930s,⁴³ has become one of the major research lines in the broad field of

hybrid materials synthesis.^{22,31,43–57} Organic molecules or monomers embedded in sol–gel matrices are common examples that could present a large diversity in their structures and final properties leading to many multifunctional materials. Polymers filled with inorganic clusters, organogels, and biological-based hybrid materials are other extended examples of class I hybrids.^{7,13,24,40,43,45,51–53,58–60} In the case of Class II hybrid materials, covalent or ion-covalent bonds are present between the organic and inorganic phases.^{20,46} In this sense, the grafting methodology, appears as a common strategy to form class II hybrid materials.^{13,14,43,44,53,58,61–64} This method, sometimes applied as a postsynthetic step, normally implies the attachment of functional organic molecules on the surface of inorganic moieties (type I–O), such as silica, titania, other metal oxides, and/or carbon surfaces.^{3,23,65,66} Sol–gel is again one of the most used as a suitable methodology for the preparation of this class of materials, with the development of hybrid materials from polyfunctional alkoxy silanes a typical example for obtaining a wide range of functional materials due to their high versatility.^{67–69} However, electrochemical grafting using aryl diazonium salts is the most used in the case of carbonaceous matrices. This method is based on the electrochemical reduction of diazonium salts, which decompose into radicals and nitrogen gas, giving a direct C–C bond.^{64,70} Other typical methods commonly used to prepare type II hybrid materials are self-assembly synthesis,^{13,36,41} template-assisted synthesis,^{24,63,71} hydrothermal,^{12,57,72,73} or layer-by-layer deposition.^{49,74,75}

The second criterion for the classification of hybrid materials focuses on the nature of the dominating structural matrix component and the one that is hosted. According to this classification, hybrid materials can be divided into two main groups: organic–inorganic (OI), when the matrix is an organic phase, and inorganic–organic (IO) hybrids, when there is an inorganic host where organic guests are integrated.²⁵ It should be noted that in a broad sense, combinations of dissimilar inorganics could also be considered as hybrid materials, which in that case could be classified as inorganic–inorganic hybrids

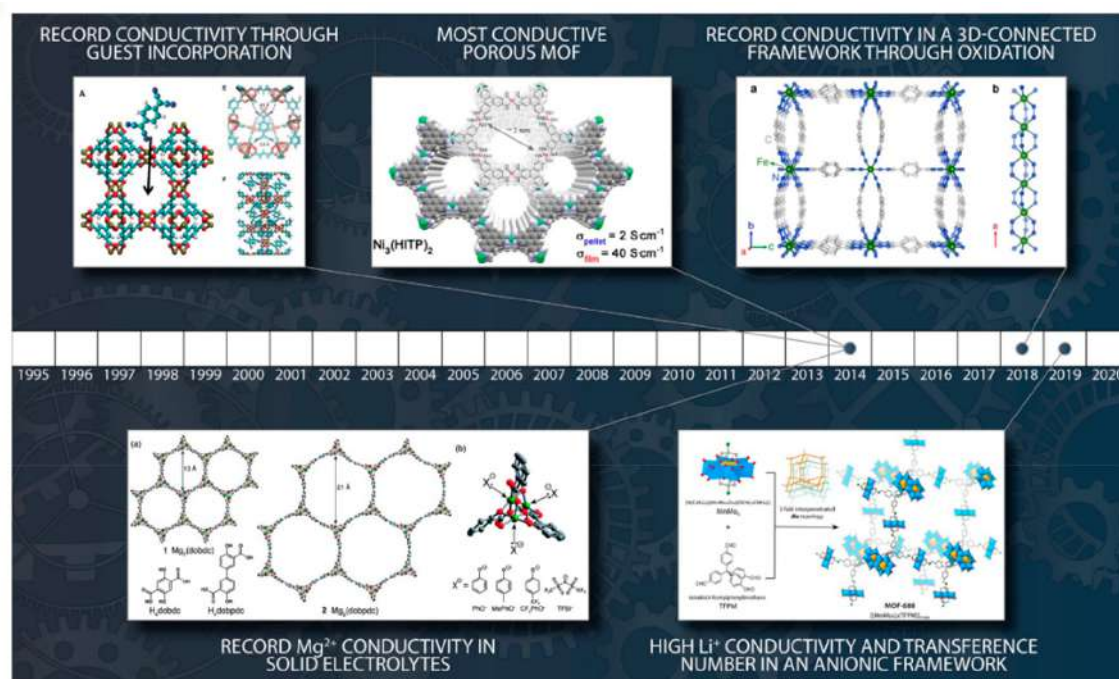


Figure 5. Examples of metal–organic framework hybrid materials (MOFs) with groundbreaking conducting properties (from REF Freund et al. *Angew Chem* 2021). Intrinsically hybrid materials, MOFs have grown into a category on their own and have led to the development of hybrids for which they act as the host framework, even leading to MOF-COF hybrids. Reprinted with permission from ref 85. Copyright 2021 John Wiley & Sons.

moieties being orderly and controllably separated by organic spacers in crystalline phases with controlled porosity and properties.^{13,27–30,79–85} This “reticular chemistry” approach allows a rational design method for creating various metal–organic frameworks and covalent organic framework (COF) structures.^{80,85,86} The main idea of reticular design is to connect organic and inorganic molecular building units with strong directional chemical bonds to form stable crystalline extended structures. A significant advantage of this method is the ability to control the pore size of MOFs and COFs by changing the length of the organic linkers used without changing the framework’s underlying topology. The past decades have seen immense possibilities of reticular design resulting in a wide range of MOF (>100 000) and COF (>570) structures that have unique properties (Figure 5).^{85,87} The great tunability of MOFs and COFs allows the design and synthesis of materials with a huge variety of properties, making MOFs and COFs a great solution for a large variety of applications such as gas storage and separation, vapor sorption, catalysis, biomedical applications, chemical sensing, and electronic and ionic conduction (such as electrode development).

Perovskite photovoltaics is another example of what began as an incipient family of hybrid materials^{1,88} then turned into a whole new realm, with lead perovskites taking the central role and lead-free derivatives trying to circumvent the problems associated with toxic lead and trying to fulfill the safe-by-design guidelines.⁸⁹

Regarding the methods of synthesis of hybrid materials, they are as varied and extensive as the chemistries of their components. Table 1 is an attempt to summarize this extensive palette and represents the strong rooting of hybrid materials in many fields of science and their huge potential to affect a vast variety of applications.

3. STRUCTURE–PROPERTY RELATIONSHIPS

Composition is obviously important in the design of hybrid materials, but the structure is equally crucial. Indeed, how structure determines properties is, in itself, a fascinating topic. From molecular chemistry to materials science, all hierarchical levels of structure have paramount importance in determining physical and chemical properties. Hybrid materials are no exception, of course. Furthermore, their multimaterial nature provides opportunities for the design of sophisticated nanostructures and molecular architectures leading to the fine control of their properties. In this section, we have selected a few case studies concerning different structures for a variety of properties and applications.

The electrochemical properties of electrodes from supercapacitors and batteries are a fitting example of a hybrid material in which the structure is a key factor. Current electrodes usually are composed of the active materials (which store energy), a conductive component, and a binder. How they are combined has evolved from the empirical simple mixture in the past to more sophisticated approaches that take into account the critical role that hierarchical structures generated in the electrode have on the electrode performance.^{34,122} Most active materials are insulators or have low electrical conductivity to work as electrodes on their own. That is why a conductive component is needed, which must generate a percolation network allowing an efficient transport of electrons from the current collector to all of the particles of active material. However, the combined structures of each of the components should be optimized. Figure 6 illustrates the importance of a properly structured composite. Thus, the top images show a real SEM image and a schematic drawing of a suboptimal combination of the three elements (active conducting and binding components). On the other hand, the bottom images in Figure 6 show electroactive particles

Table 1. Summary of the Different Synthetic Methods for Obtaining Hybrid Materials

Method	Description	Advantages	Disadvantages	Applications of interest	ref
Sol-gel	This technique involves the hydrolysis and condensation of precursor molecules, usually metal alkoxides, to form a gel-like material that can be further processed to obtain the desired hybrid structure	<ul style="list-style-type: none"> Versatile, and simple Cost-effective and scalable Control over the composition, morphology, and structure Suitability for creating complex structures Enable the synthesis of organic-inorganic composites and functional coatings 	<ul style="list-style-type: none"> Time-consuming processing Critical selection of precursors and parameters Parameter-sensitive (pH, temperature, solvent composition...) Scalability accessible but challenging Shrinkage and cracking upon drying Sensitive to contaminations 	<ul style="list-style-type: none"> Optics Electronics Catalysis Energy storage Sensing Biomedical engineering Drug delivery 	7, 12–14, 22, 31, 43–54, 57, 90–96
Self-assembly	This technique relies on the spontaneous organization of molecules or nanoscale building blocks into ordered structures driven by noncovalent interactions	<ul style="list-style-type: none"> Versatile Allow combining different components with complementary properties. Well-defined structures and functionalities can be achieved. Final hybrid material properties and functionalities can be tailored to address a specific application 	<ul style="list-style-type: none"> High influence of reaction parameters that might limit the control over the assembly. Sensitivity to environmental factors. Susceptible to contamination and purity issues. Reproducibility Challenges Limited scalability Limited Material Compatibility 	<ul style="list-style-type: none"> Biomedical imaging Tissue engineering Sensors/biosensors Optoelectronics Coatings Energy storage and fuel cells 	12, 13, 24, 36–42, 54
Layer-by-layer (LbL) assembly	This technique is based on sequential adsorption of alternating layers of dissimilar materials onto a substrate, resulting in a multilayered material. Each deposition step is followed by rinsing to remove any unbound or loosely attached material.	<ul style="list-style-type: none"> Allows precise control over film thickness Precise control of the composition and functionality Integration of diverse components with complementary synergistic properties. Multilayer films, coatings, or complex assemblies can be obtained. 	<ul style="list-style-type: none"> Time-consuming and labor-intensive process Limited thickness and scalability Challenging reproducibility and uniformity between layers Interlayer diffusion between layers. Delamination issues (weak interlayer adhesion) Incompatibilities between materials. Its proper selection is crucial. Difficulties to control the reaction. Need proper optimization of the conditions and concentrations. Polydispersity issues Compatibility between monomers and precursors on the hybrid material formation. Reproducibility and scalability issues 	<ul style="list-style-type: none"> Coatings Sensors Drug delivery systems Tissue engineering scaffolds Optoelectronic devices Piezoelectrics 	12–14, 41, 75, 97
In-situ polymerization	This method for the preparation of hybrid materials is based on the simultaneous polymerization of monomers and the formation of inorganic materials, resulting in a covalent interaction between organic and inorganic components.	<ul style="list-style-type: none"> Control over the composition Controlled morphology and structure. Enhanced mechanical properties. Versatile: varied materials can be prepared by choosing the correct precursors. Precise control of the size, shape, porosity, and composition 	<ul style="list-style-type: none"> Template removal usually may require harsh conditions or time-consuming processes. 	<ul style="list-style-type: none"> Catalysis Energy Storage Sensing Biomedical Engineering Catalysis 	12, 13, 38, 44, 59, 74, 98–100
Template assisted synthesis	The preparation of the material is conducted by using a template or scaffold that acts as a guide for the formation of the desired nanostructure.				13, 24, 29, 63, 71, 101–103

Table 1. continued

Method	Description	Advantages	Disadvantages	Applications of interest	ref
Solvothermal/hydrothermal synthesis	These two methods are based on high temperature and pressure in aqueous (hydrothermal) or organic solvents (solvothermal) to promote the nucleation and growth of the nanomaterials.	<ul style="list-style-type: none"> • Various templates can be employed such as porous materials, biological templates, sacrificial templates, and self-assembled nanostructures. • Ability to create complex structures such as hierarchical nanostructures. • Versatile <ul style="list-style-type: none"> • Well-defined hybrid structures • Controlled composition and morphology • Defined crystallinity • Can be combined with other methods • Scaling-up technique 	<ul style="list-style-type: none"> • Limitate to the suitability and availability of the templates. • Additional post-treatment steps might be required for more precise control. • Low scalability to large production. • Specialized reaction vessels are required to work with the high pressure and temperatures required. • Limited solvent compatibility • Challenging control over reaction kinetics. • Formation of undesired phases or byproducts. • Post-treatment steps might be needed. 	<ul style="list-style-type: none"> • Energy storage • Sensors • Electronics • Biomedical • Energy storage and fuel-cells • Catalysis • Environmental remediation • Photovoltaics • Optoelectronics • Sensing • Biomedical • Catalysis 	12, 57, 64, 72, 73, 103–105
Microwave-assisted synthesis	Normally related to solvothermal synthesis. It reduces the time to a few minutes and uses a lower temperature.	<ul style="list-style-type: none"> • Fast • Low energy consumption • Can significantly alter the reaction kinetics compared to conventional heating methods, reducing the energy barrier in the formation of nanomaterials. 	<ul style="list-style-type: none"> • Specialized equipment is needed. Limited applicability. • Different microwave absorption and heating selectivity of the materials may affect the homogeneity. • Can induce material degradation. 	<ul style="list-style-type: none"> • Energy storage and fuel-cells • Sensing and detectors • Photovoltaics • Environmental remediation • Optoelectronic • Biomedical • Coatings • Energy storage 	12, 65
Atomic layer deposition (ALD)	Thin film deposition technique that utilizes self-limiting surface reactions, enabling to control of the growth of thin films with atomic precision.	<ul style="list-style-type: none"> • High precision and accurate thickness. • Nonsolution-limited method (solubility, temperatures...) • Hybrid thin film deposition by sequentially reacting different precursor gases. • Controlled composition 	<ul style="list-style-type: none"> • Complex Process and Equipment • Limited scale-up potential • Limited deposition to solid substrates. • Limited material compatibility with the technique • Slow deposition rates • Limited film thickness 	<ul style="list-style-type: none"> • Optoelectronics • Microsystems • Sensors/biosensors • Microelectronics • Optoelectronics 	12, 77, 106, 107
Chemical vapor deposition (CVD)	Based on the decomposition of precursor molecules in the vapor phase to deposit thin films or nanomaterials onto a substrate.	<ul style="list-style-type: none"> • Synthesis of high-quality nanomaterials • High variety of nanomaterials can be prepared by choosing the correct precursor gas. 	<ul style="list-style-type: none"> • High-temperature requirement 	<ul style="list-style-type: none"> • Microelectronics • Optoelectronics 	12, 13, 107, 108

Table 1. continued

Method	Description	Advantages	Disadvantages	Applications of interest	ref
Chemical bath deposition	A solution-based technique used to deposit thin films or nanoparticles onto a substrate, like CVD, but it involves the reaction of precursor solutions in a chemical bath under controlled conditions.	<ul style="list-style-type: none"> Precise control over composition and structure Accurate thickness and crystallinity Simple and low cost By manipulating the bath composition and reaction parameters different hybrid materials with desired compositions and structures can be prepared. Ability to coat complex-shaped substrates Controlled thickness and composition 	<ul style="list-style-type: none"> Limited uniformity and controlled deposition for large-area substrates Limited to planar substrates. Equipment Complexity and Cost. Slow deposition rate Lack of precise control. Temperature, pH, and concentrations significantly influence the deposition rate and quality. Sensitive to contaminants Limited for specific materials Low crystallinity Postdeposition treatments might be needed. 	<ul style="list-style-type: none"> Gas sensors Catalysis Biomedical Coatings/barriers Energy storage Flexible electronics Optics Sensors Energy storage and harvesting Coatings/barriers 	13, 108, 109
Electrochemical deposition/electrodeposition	This technique uses an electric current to deposit metals or other materials onto a conductive substrate from an electrolytic solution.	<ul style="list-style-type: none"> Versatility Deposition of different materials such as metals, oxides, polymers, or organic molecules (grafting) Control over the deposition process and rate Tailored properties 	<ul style="list-style-type: none"> Limited material compatibility Electrode substrate dependency. The substrate can affect the final properties. Limited control over the thickness and uniformity Proned to pore formation cracks, or dendritic structures in the deposited films Scale-up challenges. 	<ul style="list-style-type: none"> Coatings Optoelectronics Electrocatalysis Energy storage 	13, 29, 42
Coprecipitation method	In this method, hybrid nanomaterials are prepared by simultaneous precipitation of multiple components from a solution or a mixture of precursors.	<ul style="list-style-type: none"> Simplicity Cost-effective Intimate combination of the hybrid material precursors. Enhanced properties in the final hybrid structure. Acceleration of the synthesis Homogeneous distribution of the different components Fine particle size control Morphology is controlled by adjusting the reaction conditions. Reduced temperature synthesis (compared with other methods) 	<ul style="list-style-type: none"> Lack of Control over Particle Size and Morphology. Limited control over composition; heterogeneity issues Formation of agglomerates Prone to form impurities Limited scalability Parameter sensitivity Formation of undesired byproducts Potential material degradation of highly sensitive precursors 	<ul style="list-style-type: none"> Catalysis Sensors Drug delivery Environmental remediation Catalysis Functional coatings Sensing Biomedical applications Energy storage 	12, 110–113
Sonochemical synthesis	This method uses high-frequency sound waves (typically above 20 kHz) in a liquid medium containing precursor materials. The intense acoustic cavitation created by the ultrasound waves generates localized high temperatures and pressures, resulting in various physical and chemical effects that facilitate the synthesis of hybrid materials.				14, 114, 115

Table 1. continued

Method	Description	Advantages	Disadvantages	Applications of interest	ref
Electrospinning/ electrospray	Electrospinning and electrospray techniques use an electric field to produce polymeric fibers (electrospinning) or particles (electrospray) with diameters and sizes in the nanometer range, deposited onto a conductive substrate.	<ul style="list-style-type: none"> Versatile: different combinations can be used to produce hybrid materials (such as nanoparticles, polymers, carbon materials, and biomolecules...) Tuneability and versatility 	<ul style="list-style-type: none"> High dependence on the environmental conditions (temperature and humidity) to control the fiber or particle morphology. Optimal parameter dependence 	<ul style="list-style-type: none"> Tissue engineering Environmental remediation 	12, 13, 57, 116–118
Spray pyrolysis	This method involves the atomization of precursor solutions into fine droplets and the subsequent thermal decomposition of the droplets to form solid particles or thin films. This technique allows for the synthesis of hybrid materials by incorporating multiple precursors in the spray solution	<ul style="list-style-type: none"> Allow the incorporation into the polymer matrix of other nanomaterials such as metallic and ceramic nanoparticles, nanocarbon, and/or MOFs Scaling-up technique 	<ul style="list-style-type: none"> Prone to aggregation and to limited production rate for electrospray Postprocessing may be required. 	<ul style="list-style-type: none"> Energy storage Drug delivery Sensors Catalysis 	12, 14, 119, 120
Mechano-chemical synthesis	Also known as ball milling, it involves the use of mechanical force to induce chemical reactions and synthesis of nanomaterials.	<ul style="list-style-type: none"> Controlled composition and morphology adjusting the process parameters (temperature, flow...) Simplicity Versatile Wide range of hybrid materials can be produced. 	<ul style="list-style-type: none"> High influence of process parameters Low control over particle size Aggregation and coalescence issues Challenging scale-up Energy consumption Additional post-treatment steps might be required. Time-consuming process Slow reaction rates Limited scope of reactions 	<ul style="list-style-type: none"> Energy storage Thin films fabrication Sensors Catalysis Energy Storage Environmental remediation Pharmaceutical/Drug delivery 	12, 64, 121
Microemulsion and reverse micelle techniques	This technique involves surfactants and organic solvents to form stable nanoscale droplets or micelles in a continuous phase, allowing the formation of different nanomaterials in the confined space inside the droplets.	<ul style="list-style-type: none"> Simple and low-cost Scalable High yield Solvent-free processing Control over the composition Can be also liquid or ultrasonic-assisted. <p>Simple and easy</p> <p>A wide range of materials can be used from hydrophobic to hydrophilic, including organic and inorganic compounds, polymers, and biomolecules.</p> <p>Controlled particle size and morphology</p> <p>Unique microenvironments can be created in confined spaces.</p> <p>High homogeneity and stability.</p> <p>Controlled release and delivery.</p>	<ul style="list-style-type: none"> Impurities might be obtained from the jars and grinding balls. Broad particle size distribution Additional post-treatment steps Limited control over particle size distribution and morphology Limited material compatibility Limited loading capacity Affected by diffusion rates Post-treatment needed Environmental impact by surfactants 	<ul style="list-style-type: none"> Drug delivery Cosmetics Biomedical applications Catalysis 	13, 44, 93, 95

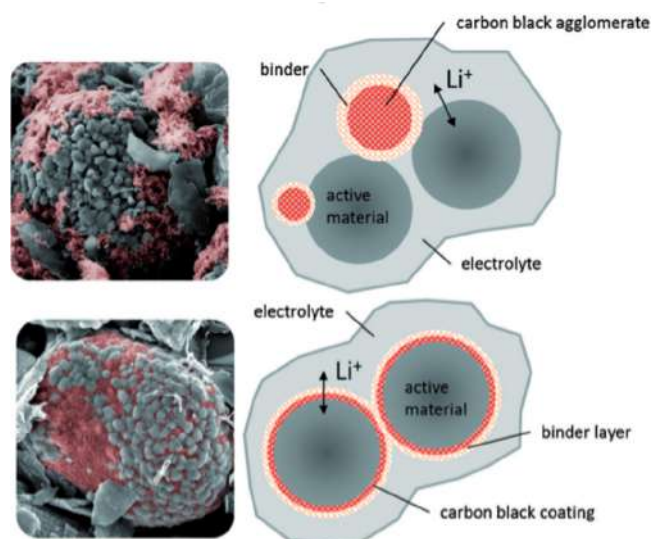


Figure 6. An example of the importance of microstructure in the field of hybrid nanocomposite electrodes: The top images show a real SEM photograph and a schematic rendering of the improper agglomeration of carbon and binder. The bottom images represent the corresponding SEM and diagram of a sample electrode with a proper coating of the active material, resulting in improved performance. Reprinted with permission from ref 123. Copyright 2015 John Wiley & Sons.

properly coated with conducting carbon and the binder uniformly distributed but without blocking percolation pathways, which correspond to an optimized, properly working electrode.¹²³

Encapsulation is another example of a hybrid material in which the structure is a key feature. Some applications use hybridized inorganic nanoparticles as active pharmaceutical ingredients, vectors, or enablers. Others employ them for *in vitro* diagnosis in lateral flow devices (immunochromatography) or in delivery systems (drug delivery, implantable biomaterials, and vaccine adjuvants). The field of cosmetics and controlled release of “active ingredients” is the target of these new hybrid compounds; they are especially useful for skin care and protection applications. As an example, silica microcapsules can reduce skin contact with harmful organic UV filters by their encapsulation in microcapsules, which is important because these chemicals can produce free radicals that damage DNA. These “UV-pearls” can be mixed with a suitable cosmetic vehicle to achieve high sun protection factors (SPFs), while improving the safety profile as the penetration of the UV filters is significantly reduced. Some companies have already used these “UV-pearls” for sunscreens and daily wear cosmetics (Figure 7).¹²

Structure is also key in biomedical applications in general, polymer bioconjugates, and the conjugation of biomolecules with synthetic polymers is a smart solution to get a hybrid material with advanced properties. In this field, the influence of polymer topology on the properties and applications of polymer bioconjugates is significant. Polymer bioconjugates are hybrid biomacromolecules that combine synthetic polymers with various biomolecules. Branched polymers, such as brushlike, hyperbranched, and dendritic polymers, have been widely used for biomedical applications due to their unique features compared to linear polymers. Therefore, the synthesis of branched polymer bioconjugates has become a

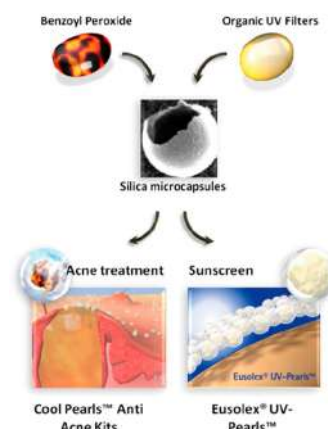


Figure 7. Encapsulation as a primary structuration method in hybrids. From drugs for acne treatment to sunscreens, silica microcapsules provide optimal delivery. Reprinted with permission from ref 12. Copyright 2009 Royal Society of Chemistry.

promising research area to obtain biohybrid materials with enhanced stability and prolonged circulation times *in vivo*.

The design of polymer bioconjugates depends on a range of factors, such as the site-specific conjugation chemistry; the size, distribution, topology, and function of polymers; and the architecture of bioconjugates. These factors affect the higher-ordered assemblies and hierarchical structures of polymer bioconjugates in solution, in bulk, and on surfaces. The synthetic tools and methods for creating polymer bioconjugates have been rapidly developed and improved in recent years. Moreover, the understanding of biomolecule structure and function has also been deepened, leading to novel constructs and applications in materials science.

The field of polymer bioconjugates is constantly evolving and expanding. The synthetic chemistry of macromolecules offers a wide range of possibilities that surpass those found in nature. However, nature also provides inspiration and guidance for creating complex and functional systems that can communicate with and regulate themselves. The future of polymer bioconjugates may lie in establishing the molecular principles of how these macromolecules can be customized and integrated into artificial environments. The potential applications of these biohybrid materials are enormous and diverse.^{60,124–126}

Finally, biodetection applications also require a tailor-made adjustment of the physical, chemical, and structural properties of nanomaterials. For example, metal nanoparticles and quantum dots can be tuned to have different optical properties, such as emission, absorption, and scattering, by changing their size, shape, and composition.^{53,94} This allows for the detection of multiple analytes simultaneously using different colors of light; nanowires and nanotubes can also be modified to have different electrical properties, such as conductivity and resistance, depending on the presence of target analytes. Furthermore, nanomaterials can be functionalized with biomolecules or small molecules to enhance their specificity and affinity for various targets. These advances in nanotechnology enable the fabrication of nanoscale arrays of sensors on surfaces. One example of such a sensor is the colorimetric assay based on gold nanoparticles that change color when they aggregate in response to DNA hybridization. The aggregation of gold nanoparticles alters their surface plasmon resonance and scattering properties, resulting in a

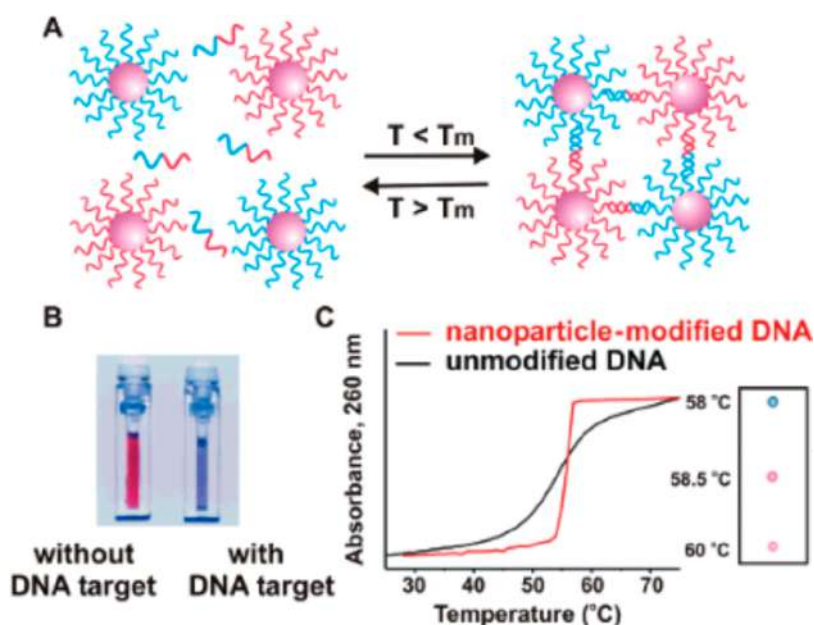


Figure 8. Oligonucleotide-functionalized gold nanoparticles aggregate in the presence of complementary target DNA (A), resulting in a color change from red to blue (B), which can be monitored by UV-vis spectroscopy (C). Reprinted with permission from ref 127. Copyright 2005 American Chemical Society.

visible color change from red to blue. This assay can be used as a simple and rapid test for nucleic acid detection by spotting the solution onto a white support (Figure 8).¹²⁷

4. APPLICATIONS

The variety of compositions and chemical structures that arise from all the possible combinations of organic and inorganic components makes hybrid materials not only suitable but also an excellent option for a wide range of applications. On top of that, the synthesis method can be tailored to achieve specific properties, providing a powerful tool to address current technological problems.²⁰ It is evident that the applications of hybrid materials are numerous and diverse, spreading through many fields, including energy storage, catalysis, sensing, photonics, and biomedicine. However, within each of these fields, the applications of hybrid materials are vast, and describing them all in one review has become an impossible task. Again, it is inevitable to think about the tree metaphor (Figure 1), in this case a branching tree, focusing now on the different main branches and individual leaves, each with its numerous nerves. Thankfully, it is common now to find reviews dealing with hybrid materials that find application in only one specific area, and in this section, we will provide the readers with a summary of these articles attempting to assist them in their journey through the world of hybrid materials and their applications. In this regard, those looking for a first glance at the properties and fields of application, should refer in a first instance to general reviews^{3,12,20,95,128} which usually include a brief description of possible uses of hybrid materials or current technological advances. Based on these reviews, the myriad of applications found for this kind of material have been grouped as follows:

4.1. Optics and Photonics. This is one of the first fields in which hybrid materials found application. By doping inorganic materials like glass, clays, silica, and zirconia with organic dyes, it has been possible to improve the photostability of the chromophore compound, an approach trying to reproduce one

of the oldest hybrid materials, Maya blue. Since these first studies, more sophisticated developments seeking to achieve different optical properties have been developed. For example, polymer-based hybrids containing thin films of Ti or Zr metal oxides, show refractive indexes higher than those of the individual components.³ Furthermore, fast and reversible photochromic materials can be obtained by limiting the organic–inorganic phase interactions by simply modifying the dye-doping procedure.⁵³ The list of optical devices that can be produced with hybrid materials is, as expected, quite large, but some examples include high or low refractive-index materials, waveguide materials, photochromic⁹⁴ and electrochromic materials, nonlinear optical materials,⁵³ photodetectors, and decorative coatings. The reader interested in this particular topic is encouraged to consult the thorough review by Lebeau and Innocenzi,⁵³ where a comprehensive list of the applications and references can be found. It is also worth mentioning that this review is mainly focused on sol–gel materials. However, it does provide a good idea of the potential of hybrid materials in the fields of optics and photonics.

4.2. Biomedical Applications. The versatility of hybrid materials makes them suitable for several applications within this field, like tissue engineering, drug delivery, dentistry, and bioimaging. Once more the idea is to combine the properties of organic and inorganic phases, keeping in mind that the latter in this case must be nontoxic and biocompatible. This is one of the reasons why silica-based hybrids are currently the most popular ones, especially since it has been approved by the FDA for human trials.⁶⁶ Silica nanoparticles provide a stable platform which can be easily functionalized with different materials, biomolecules, and targeting ligands, thanks to the presence of Si–OH groups on its surface. The interaction between silica and the organic moieties can be either weak (drug delivery) or strong (tissue engineering) depending on the requirements of the materials.¹²⁹ Moreover, porous structured silica particles (MCM-41 or SBA-15 type)⁵⁴ can be used as carriers of bioactive molecules entrapped within the

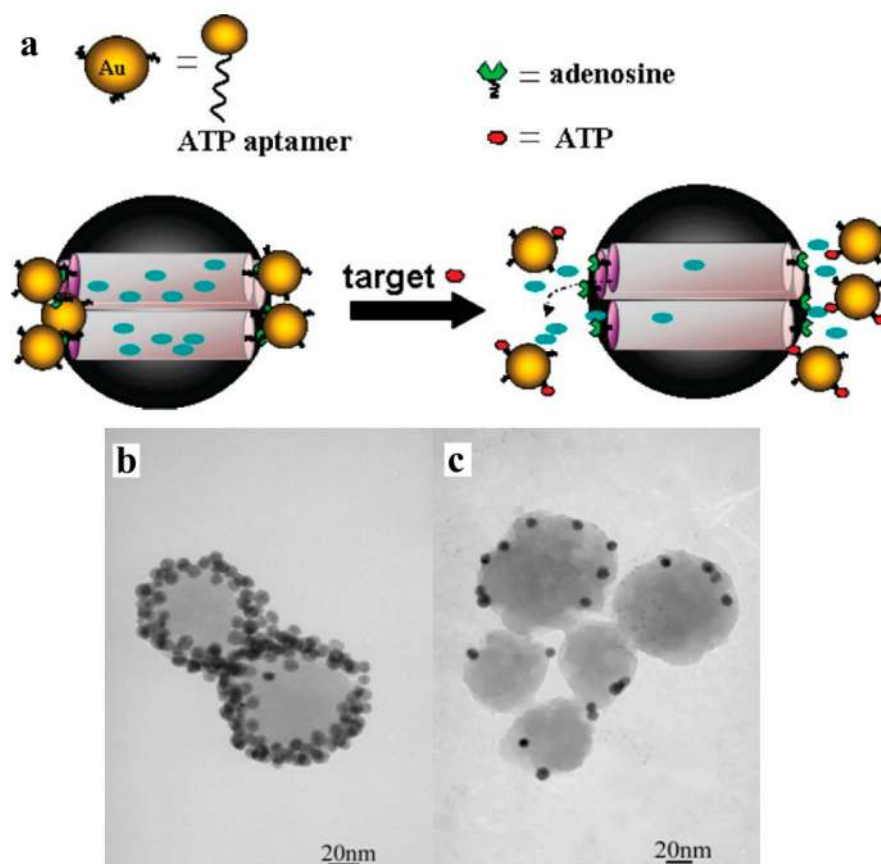


Figure 9. (a) Scheme showing working principle of Au nanoparticle capped-mesoporous silica as responsive controlled drug delivery systems. TEM images of these particles in the (b) before and (c) after being exposed to ATP rich environment. Adapted with permission from ref 131. Copyright 2011 American Chemical Society

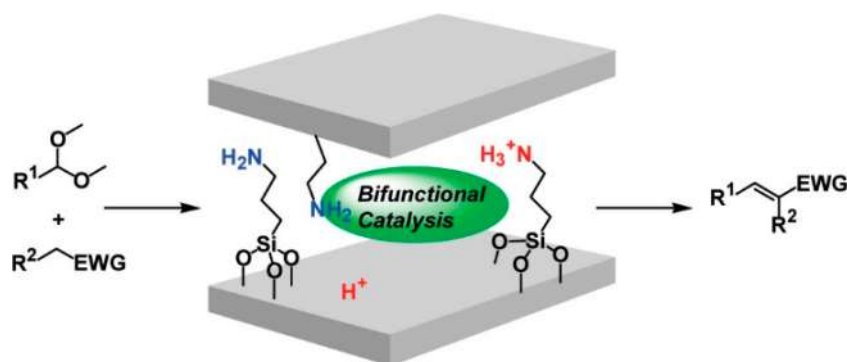


Figure 10. Acidic montmorillonite-immobilized primary amines as acid–base bifunctional catalysts for cascade reaction. Reprinted with permission from ref 15. Copyright 2009 American Chemical Society.

pores by “gatekeeper” molecules, polymers, or even metal/metal oxide nanoparticles. These are usually designed to respond to either external triggers such as temperature, light, magnetic field, ultrasound, and electricity or internal triggers such as glucose, enzymatic activity, pH, ATP, and glutathione (Figure 9), which makes it possible to target only affected areas. Even more complex silica-based materials, containing both therapeutic and imaging agents,^{66,128} have been devised for the real-time monitoring of drug release and therapeutic response (theragnostic systems). Silica-based 3D scaffolds have also been proposed for bone tissue engineering since it is possible to graft osteoinductive agents which act as attracting signals for bone cells promoting tissue growth and regener-

ation. Other types of materials that are attracting interest in this field are metal–organic frameworks (MOFs)^{79,128} which can be easily combined with biopolymers to comply with biocompatibility requirements.¹³⁰

4.3. Catalysis and Electrocatalysis. Hybrid materials were first used in catalysis to increase the stability of organic or organometallic homogeneous catalysts. In this type of material, the organic phase was retained within the structure of the inorganic phase only by weak interactions (van der Waals, hydrogen bonds, or electrostatic), falling within the class I classification proposed by Sánchez and Ribot.³³ Even though leaching, pore blocking, and subsequent deactivation of the catalyst are still major issues of these hybrids, this approach

solves the most important problems of homogeneous catalysis such as product separation or recovery and catalyst reusing. Organic molecules occluded within the cavities of zeolites are one example of catalysts that have been prepared in this way. A second stage in the development of hybrid catalysts was mainly focused on overcoming the main drawbacks of Class I type materials by establishing a covalent bond between both phases (Class II). This ensures not only that active organic molecules are stable (no leaching) but also that they are homogeneously distributed so the whole material (pores and cavities) can be fully exploited. The latter is also true since the organic counterpart is now confined within the walls of the solid and does not block the internal channels of the solid, allowing reactants to diffuse inside. Silica and silica–alumina have been frequently employed as inorganic supports due to their large specific area and high number of reactive sites, which can be used to anchor the organic compounds. However, layered oxides have also been used to produce bifunctional acid–base catalysts (Figure 10). A final comment must be made about a third kind of hybrid materials that has gained popularity in this field, namely, organic modified/functionalized metal and metal oxide nanoparticles.^{132,133} This is an emerging area that uses surface functionalization to manipulate the catalytic properties of nanoparticles. As an example, it has been reported that modification of Ru particles with ethylenediaminetetraacetic acid (EDTA) changes the selectivity of the catalyst in Fischer–Tropsch synthesis.¹³⁴

4.4. Energy Storage and Conversion. In the field of energy storage, hybrid materials have attracted a lot of attention since combination, for example, of carbon materials with pseudocapacitive materials (metal transition oxides or conductive polymers) can help overcome the limitations they show individually and boost the performance of supercapacitors.¹⁷ Going a step further, faradaic materials could be incorporated in the network of capacitive-like materials to prepare electrodes with a hybrid electrochemical response. The latter provides a way to close the gap between batteries and supercapacitors in terms of the energy and power density. For energy storage, the structure of the materials (see the previous section) plays a fundamental role in the performance of the resulting devices. For example, decreasing the size up to the nanoscale can even lead to capacitive-like behavior of materials commonly known as “faradaic”. The possible combinations are infinite, and even multiple hybridizations (carbon/metal oxide/conductive polymer) have been considered.¹³⁵ On this topic the reader can find specific reviews such as those by Gómez-Romero et al.^{69,136} dealing with polymer–metal oxides (including polyoxometalates) hybrids or that of Reddy et al. which focuses mainly on CNT-based hybrids⁷¹ for energy storage applications. Solar energy harvesting is another field that is taking advantage of the benefits of hybridization. Organic–inorganic perovskites are materials with the typical chemical formula ABX_3 , where B is a divalent cation and X a halogen anion, but unlike traditional perovskites A is an organic cation (e.g., methylammonium).¹³⁷ These materials have gained popularity due to their ease of preparation, low cost, and high efficiency. Also, the right combination of metal and organic cations as well as halogen anion can provide the desired bandgap.¹³⁸ Although these materials show great potential, commercialization is just starting and other aspects need to be addressed.

4.5. Sensing. The field of sensors is overly broad, since they can be used to detect gases, chemical species, biomarkers

in biologic systems, humidity, mechanical deformations (strain or pressure), temperature, or UV-radiation. This in turn means that the composition of hybrid materials developed for sensing applications can be diverse. Thus, for those interested in this topic, the review by Wang et al.¹³⁹ is a good starting point, where preparation methods, sensing configuration, shape of sensor, and an overview with examples of materials used in diverse kinds of sensors is provided. According to these authors, the most general approach consists of protecting the organic sensing molecules within the inorganic matrix. However, as observed for other applications, hybrids can be designed in such a way that there is a synergistic effect between both components, enhancing the performance when compared to individual materials (Figure 11). Carbon-based hybrids, e.g.,

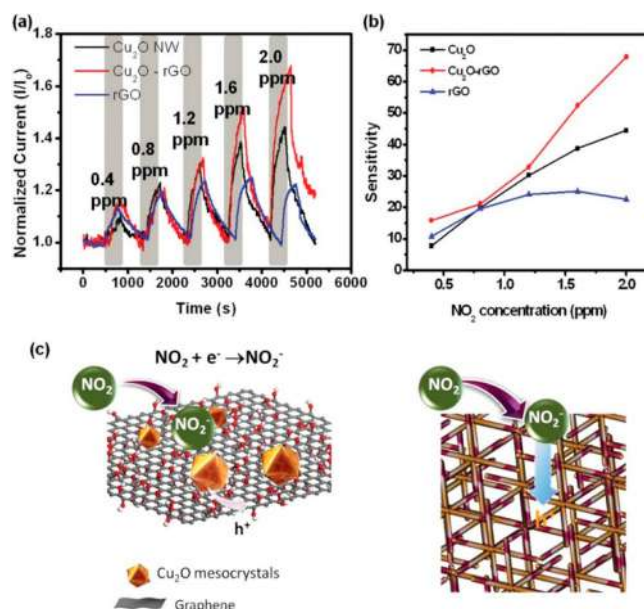


Figure 11. (a) Dynamic response of Cu₂O nanowires, rGO-Cu₂O, and rGO materials under increasing NO₂ concentrations, showing the synergistic effect of both phases. (b) Sensitivities of NO₂ sensors for the three devices. (c) Mechanism of NO₂ sensing of rGO-Cu₂O. Reprinted with permission from ref 32. Copyright 2012 American Chemical Society.

graphene¹⁴⁰ and CNT,¹⁴¹ have been extensively considered for this purpose to take advantage of the high conductivity, specific area, and good thermal stability. Besides, the surface of these nanocarbons is already reactive, so they can act as sensors. In this case, hybridization with either polymers or metal/metal oxide nanoparticles can further enhance their sensitivity. Photofunctional hybrid sensors have been prepared by encapsulation of metal ions or nanoparticles within the structure of MOF. These materials have proved to be efficient in the luminescent detection of a wide range of biomarkers.⁷⁹ Another interesting review in this field deals with the combination of phthalocyanines and metal nanoparticles to obtain hybrid materials with excellent properties for chemiresistive and electrochemical sensing.¹⁴² Finally, it is worth making a short comment on the development of flexible sensors. This emerging field has many interesting applications, such as electronic skin, medical electronics, environmental detection, and wearable devices, which could benefit from the properties of hybrid materials.¹⁴³ For example, it is not hard to imagine how the combination of flexible but low-sensitivity

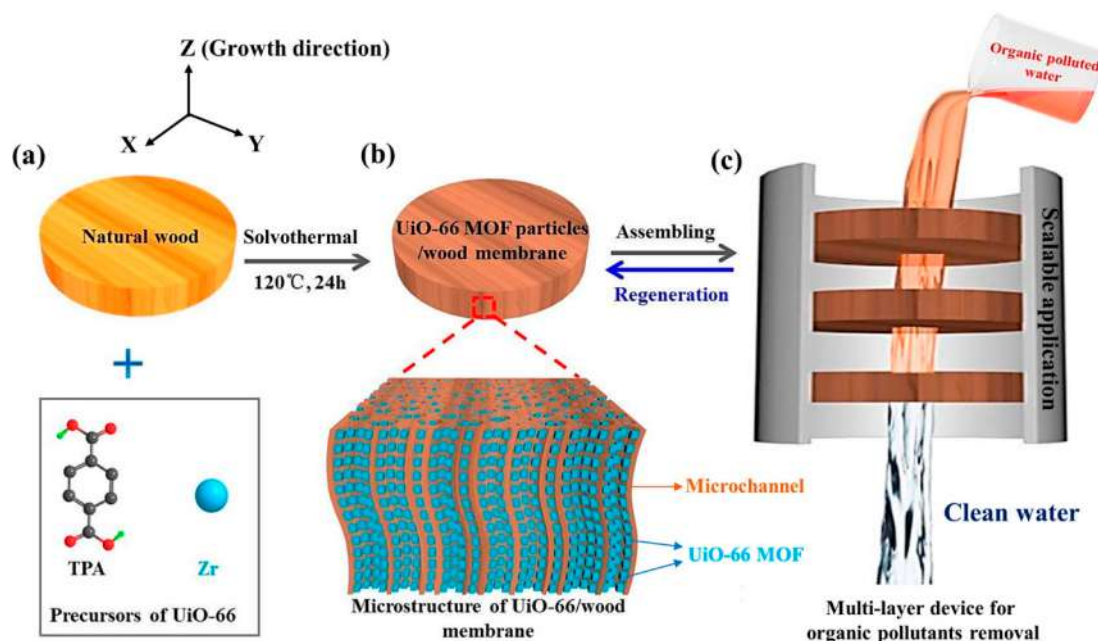


Figure 12. (a) Scheme of the in situ synthesis process and (b) of the resulting structure of a UiO-66 MOFs-wood hybrid membrane for efficient organic pollutant removal. (c) Filter built with 3 membranes for large-scale operation. Reprinted with permission from ref 104. Copyright 2019 American Chemical Society.

organic materials with inorganic semiconductors can accelerate the implementation of these devices in real-life applications. An excellent summary of the possibilities and current limitations of these devices has been provided by Ren et al.,¹⁴³ together with over 100 references for interested readers.

4.6. Electronics. The term “electronics” is quite broad, and many different applications fall within this topic, including photovoltaics, which in this paper have been discussed briefly as energy conversion materials. However, in several reviews, hybrid materials for solar cells are included in the electronic applications section, or to be more precise under the optoelectronic label. Although both classifications are correct, here we focus on other applications such as transistors, diodes, and memory devices. The reader should keep this in mind while consulting the articles cited in this section since they do not strictly follow our approach. Thus, the general review by Mir et al.¹²⁸ includes a brief introduction to the many advantages of hybrids for electronics (photovoltaics included), electrical memory materials, and flexible devices. This field is mostly dominated by two distinct types of materials: (a) conductive polymer-based hybrids with nanocrystals or nanoparticles of inorganic semiconductors or metal nanoparticles and (b) hybrid halide perovskites. Regarding the first group, the article by Reiss et al.⁵⁸ provides a complete list of interesting materials, the hybridization routes, the electronic properties of the resulting hybrids, and even the characterization techniques used to determine their physicochemical properties. In a similar way, Holder et al.¹⁴⁴ discussed the application of these materials in the field of optoelectronics, with a special emphasis on light-emitting diodes. The fundamentals aspects and synthesis methods of hybrid perovskites for several applications (transistors, memory devices, and artificial synapsis) are properly discussed by Choi et al.,¹⁴⁵ providing a great starting point to understand how these materials could revolutionize the field. Finally, we would like to mention the work by Hwang and Lee⁸⁹ which

deals only with the design of memory devices based on hybrid materials.

4.7. Environmental Remediation. The design concept of hybrid materials for the removal of hazardous pollutants spans from simple functionalized porous silica materials⁶³ to complex polymer–inorganic semiconductor photocatalysts.¹⁴⁶ Of course, the mechanisms through which these toxic chemicals are eliminated from water courses, industrial effluents, or the atmosphere (in the case of gases) are completely different. Nonetheless, the idea is always the same, i.e., to get the best of both worlds. For example, as has been discussed in previous paragraphs, porous silica materials provide a high surface area platform to which organic molecules can be easily attached. The latter can be chosen to interact with heavy metal ions or organic dyes, which will be adsorbed and fixed at the surface of the silica particles. In this way, by simply recovering the solid particles, remediation of aqueous media can be efficiently done. Using a similar concept, researchers have been working on the development of magnetic hybrid sorbents which can then be separated from the treated sample by means of a magnetic field.¹⁴⁷ Graphene, carbon nanotubes, and MOF based hybrids with iron or iron oxide as magnetic phase have been studied, among others. Three-dimensional graphene hybrid materials with nanoporous and microporous structures have been produced for water purification and environmental monitoring. These materials have been tested as filtration membranes, adsorbents, and as pollutant degradation agents.¹⁴⁸ Other promising devices in this field are MOF-based hybrid filters (Figure 12). With high flux and low-pressure drop, they show great potential for both air and water purification.¹⁴⁹

4.8. Coatings. The use of hybrid materials as protection coatings, especially for corrosion inhibition, is one of the most obvious and straightforward applications. In a sense, these materials are the evolution of traditional paints, where particles of inorganic oxides are dispersed in a polymeric matrix. With this in mind, a lot of effort has been put into the design and

preparation, mostly through sol–gel methods, of polysiloxanes coatings. One of the advantages of this kind of material is the capacity to react with the –OH groups at the surface of metallic surfaces, leading to a strong substrate–coating interaction (i.e., great adhesion).¹⁵⁰ Class II hybrids can be tuned to have high hydrophobicity, good corrosion protection (barrier effect), low dielectric constants, or good scratch resistance, fulfilling all the requirements of a corrosion mitigation material.⁵¹ However, the possibility of hosting different functionalities within the siloxane matrix widens the range of applications for hybrid materials. Self-healing, self-cleaning, antifouling, fire-retardant, and antireflective coatings have already been prepared.¹⁵¹ Regarding the suggested literature, the mini-review by Zvonkina and Soucek¹⁵¹ introduces the topic, giving some details on the different types, preparation, and applications of sol–gel hybrid materials as coatings. Moving on, in-depth information on the use of hybrid coatings for corrosion protection (and microbiologically induced corrosion) can be found in the articles by Figueira et al.^{50,51} and Al-Saadi and Raman.¹⁵⁰ Finally, the review by Zhang et al.⁹⁶ summarizes the advances and design strategies of polymer–ceramic hybrid antifouling coatings based on chemical hybridization.

4.9. Other Applications. At the beginning of this section, the multiplicity of hybrid materials was highlighted, mentioning how the latter creates an entire world of opportunities when it comes to applications. The most common ones have been discussed briefly in previous paragraphs, but it should be kept in mind that this list is surely incomplete. Being a fast-growing field, new materials are constantly being prepared, which may find application in areas that have not been considered before. While going through the literature we have come across mentions of hybrid materials being used in aerospace and automotive applications¹⁵² or as membranes for forward osmosis.⁷⁴ Moreover, biopolymer-based hybrids are currently being studied as functional food-packaging materials.¹²⁴ As a closing remark, we would like to remind readers that this is only a glimpse of the world of hybrid materials and that their applications right now are limited only by our imagination.

5. CONCLUDING REMARKS: PERSPECTIVE AND PROSPECTIVE

The classical approach to chemistry is one in which purity is a cornerstone: extraction, purification of mixtures, isolation of compounds, and only then analysis and understanding. But when it comes to the chemistry of materials, purity is not necessarily an asset. On the contrary, complexity frequently reigns even in simple single-compound materials, and of course, in hybrid materials complexity comes standard. Nevertheless, complexity is not a problem but an opportunity for the design and development of new materials with the desired properties, and all of the materials and types of materials that we have discussed in this metareview are good examples of this.

Indeed, in this article we have tried to provide a broad perspective of the expanding world of hybrid materials, from its origins to the development of new hybrid trees of knowledge, trying also to go beyond the mere enumeration of types or applications. Thus, we have strived to underscore common factors that could inspire young researchers to tackle the challenge of hybrid diversity. This approach renounces necessarily going deep into any specific field, but this is not

a problem in view of the growing number of diverging topics and reviews dealing with hybrid materials. The practitioner searching for specific and specialized knowledge will always find excellent reviews, from hybrids for energy storage to theragnostics. This metareview, in contrast, is devoted to a more general and transversal view of a field that is not a single field anymore and tries to convey the awesome feeling that comes when we realize how the chain-reaction development of a topic like that of hybrid materials has led in a few decades to a cascade of new topics and subtopics, even new fields contributing to the development of new materials and new devices for the improvement of our way of life.

A prospective analysis of the development of hybrid materials could include consideration of new design tools, new types of hybrids, and their future impacts.

New design tools should come from both synthesis and analysis. In the synthetic corner, an empirical trial-and-error conventional approach should give way to a more rational approach based on or at least supported by theoretical calculations and modeling.¹⁵³ On the analytical side, the characterization of materials has already grown into a very sophisticated field, with the instruments of nanocharacterization and the power of large installations like synchrotrons put to the service of ever more complex materials like our hybrids.

Future impacts will concern new fundamental knowledge made possible, in part, by those new tools discussed above. New knowledge, driven as always by intellectual audacity, will keep growing and contribute to the explosion of open scientific publications. But hybrid materials will certainly also have a growing impact concerning final, real-world applications. Even as the world experiences contractions of demand due to pandemics and faces a supply crisis, there is still a growing technological market, demanding more and more materials with multifunctional and tailor-made properties, creating the perfect context for hybrid materials to thrive.

Finally, concerning the types of materials that are foreseen on the horizon of hybrid materials, there are reasonable predictions, some related to their nature and some to their applications. Concerning applications, it is most likely that hybrids will get consolidated for biomedical applications and that their use will grow in energy-related applications. Concerning the nature of future hybrid materials, it is only natural that their variety will keep growing. This, together with a trend toward growing complexity of multimaterial designs, can be foreseen as a response to the growing trend in tailor-made applications. But in the field of hybrid materials, it is truer than ever that the limits in material design are constrained just by our limited imagination.

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Notes

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