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# Applications of Organic Molecular Nanostructures

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ABSTRACT: Conjugated organic molecules are interesting materials because of their structures and their electronic, electrical, magnetic, optical, biological, and chemical properties. However, researchers continue to face great challenges in the construction of well-defined organic compounds that aggregate into larger molecular materials such as nanowires, tubes, rods, particles, walls, films, and other structural arrays. Such nanoscale materials could serve as direct device components. In this Account, we describe our recent progress in the construction of nanostructures formed through the aggregation of organic conjugated molecules and in the investigation of the optical, electrical, and electronic properties that depend on the size or morphology of these nanostructures. We have designed and synthesized functional conjugated organic molecules with structural features that favor assembly into aggregate nanostructures via weak intermolecular interactions. These large-area ordered molecular aggregate nanostructures are based on a variety of simpler structures such as fullerenes, perylenes, anthracenes, porphyrins, polydiacetylenes, and their derivatives. We have developed new methods to construct these larger structures including organic vapor-solid phase reaction, natural growth, association via self-polymerization and self-organization, and a combination of self-assembly and electrochemical growth. These methods are both facile and reliable, allowing us to produce ordered and aligned aggregate nanostructures, such as large-area arrays of nanowires, nanorods, and nanotubes. In addition, we can synthesize nanoscale materials with controlled properties. Large-area ordered aggregate nanostructures exhibit interesting electrical, optical, and optoelectronic properties. We also describe the preparation of large-area aggregate nanostructures of charge transfer (CT) complexes using an organic solid-phase reaction technique. By this process, we can finely control the morphologies and sizes of the organic nanostructures on wires, tubes, and rods. Through field emission studies, we demonstrate that the films made from arrays of CT complexes are a new kind of cathode materials, and we systematically investigate the effects of size and morphology on electrical properties.Low-dimension organic/inorganic hybrid nanostructures can be used to produce new classes of organic/inorganic solid materials with properties that are not observed in either the individual nanosize components or the larger bulk materials. We developed the combined self-assembly and templating technique to construct various nanostructured arrays of organic and inorganic semiconductors. The combination of hybrid aggregate nanostructures displays distinct optical and electrical properties compared with their individual components. Such hybrid structures show promise for applications in electronics, optics, photovoltaic cells, and biology. In this Account, we aim to provide an intuition for understanding the structure-function relationships in organic molecular materials. Such principles could lead to new design concepts for the development of new nonhazardous, high-performance molecular materials on aggregate nanostructures.

KEYWORDS: organic molecular, nanostructures, applications, hybrid, nanoscale, nanotubes, design

#### I. INTRODUCTION

Molecular nanostructures with well-defined particle shape and size, such as dendrimers and dendritic structures, are of great importance in the development of novel therapeutic devices. The unique properties of dendritic macromolecules render them far more useful for biomedical applications than linear polymers. Thus, over the past three decades, sizeable contributions to the polymer-based nanocarriers have come from dendrimers that, along with their endocavities, provide a wide scope for structural modifications at the molecular level. <sup>1</sup> New nanomaterials could be

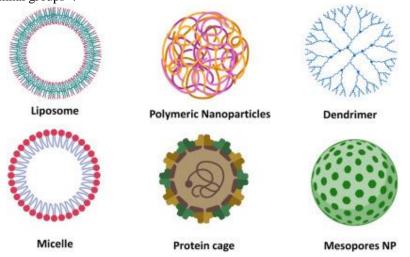


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prepared using these structures as building blocks. Nanoparticle-cored dendrimers are also versatile carriers because of their colloidal stability, tunable membrane properties, and ability to encapsulate or integrate a broad range of drugs and molecules. These hybrid nanocomposites aim to combine the advantage of stimuli-responsive dendritic coating, in order to regulate drug-release behavior under different conditions, with improvements in the biocompatibility, and in vivo half-time circulation of the inorganic nanoparticles. Assembling functional molecular building blocks on surfaces is one of the most extensively studied areas in nanoscience and nanotechonology. Through molecular self-assembly on solid surfaces, long-range ordered functional molecular nanostructures have been fabricated. Thanks to the noncovalent interactions, including hydrogen bonding, halogen bonding, van der Waals interaction, and coordination interaction,<sup>2</sup> the scale of ordered domain reaches several thousands of nanometers, with nearly no topological defect. The result is understandable from thermodynamic point of view. Since the noncovalent interactions are relatively weak, molecule self-assembly on surfaces usually proceed in a reversible manner. And thus, defects formed kinetically can be repaired through molecular reorganization, which leads to long-range well-ordered molecular nanostructures. Organic NMs with low noncovalent interactions that have been explored to model and independently assemble molecules in desirable configurations have the theoretical benefit of building blocks utilizing polymer technology at various levels and enabling the upward direction for the synthesis of organic and polymer nanostructures. For instance, self-assembling of organic moieties utilizing hydrogen bonding, electrostatic interactions, and pi-stacking would be an effective fusion strategy to the molecular nanostructure's formulation attracting special attention as a paradigm of nanotechnology, since 1990s. However, it is difficult and demanding to assemble well-defined NMs on wide areas, tune noncovalent interactions for effective integration, and establish the complexity and simulation of the self-assembly process.<sup>3</sup> This is desirable that organic biomaterials are produced in vast areas (mass production) in quite a smooth, efficient, and controllable way, and their implementations in the industrial optoelectronic devices are currently under development . Recent predictions on organic-based nanostructures, as bioactive constituents have not included the significant effort to control and customize their surface morphology. The general aims of such a chapter will be to combine scientific knowledge and implementations on organic NMs with a focus on critical analysis and preparing, processing, classification, and technologies of organic NMs, which provide new or better features, experimentation, and applications, such as drug delivery, molecular imaging, and nanoelectronics .Dendrimers are tiny and radially symmetrical molecules with a transparent, uniform, monodisperse structure, usually in a symmetrical heart, an internal shell, and an external shell. Dendrimers are hyperbranched tree-like architectural arms with engineered tailored architecture at the terminal groups <sup>4</sup>.



Polymeric NMs are divided into natural polymer-based and synthetic-based on their source of generation. This is feasible to biodegrade natural polymers into the water, CO<sub>2</sub>, and tiny inorganic molecules and to mark them as



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environmentally sustainable products, as a green resource. Varieties of different natural polymers can be generated with the desired terminal functional group through the physical and chemical processes to introduce the desired moieties. In the pharmaceutical field, natural polymer work has been performed to focus on specific natural polymers that are currently being studied, including chitosan (CS), cotton, alginate, hyaluronic acid, cellulose, and starch chondroitin sulfate. CS is highly significant, as their biological viability and biodegradability are enhanced via electrostatic interactions<sup>5</sup>. CS can bind to negatively charged proteins or plasmid deoxyribonucleic acid (DNA) and form polymer nanocomposites that preserve (encapsulate to protect) protein and DNA against enzymatic degradation, which are ideal to use them as an adjuvant or drug carrier vesicle. CS has excellent absorbability, permeability, and the ability to retain its moisture .The abundance of amino and hydroxyl groups on the backbone of CS makes chemical modification (hydroxylation, alkylation, sulfation, quaternization, acylation, and carboxymethylation) feasible<sup>6</sup>.However, derivatives of CS often have better performance than CS due to their excellent water solubility. CS products have considerable potential in the medical sector due to their significant biological characteristics, including microbial efficacy and nontoxicity.Starch is readily available in corn, bean, and potato fields; it is not toxic, lacks immunogenicity, has strong storage stability, has no medication reactions, and is generally used in pharmaceutical products. Further, starch is used as therapeutic carriers, tablet excipients, bone repair agents, and a defensive agent of blood and plasma constitutes. The microsphere surface area will increase; functional groups will be clustered and selectively adsorbed; the adsorption transition period will increase with consistency; and a slow drug-release profile will become more noticeable in starch-based microspheres<sup>7</sup>.

Alginate is a naturally occurring polysaccharide primarily produced by marine plants. Sodium, calcium, and ammonium alginate are the most marketable alginate present in the field. Alginates are grouped according to their composition and relative molecular weight, such as very low, moderate, and stable viscosity. The inconsistency in alginate performance is often a consequence of the material's surface functional variation. Many of the alginates' attributes will include effective moisture absorption, efficient and easy removal of oxygen, high oxygen permeability, biodegradability, and biocompatibility<sup>8</sup>.

Microbial hydrolysis enzymes are used to form biosynthesized polymeric NMs, such as poly-β-hydroxybutyrate (PHB), polyamino acid, and biofibers, and are formed under unfavorable growth condition (limited nutrients). Such polymers have the same properties, such as natural polymers from biodegradability, biocompatibility, and conductivity (piezoelectricity). Further, these polymers were used as tissue engineering, bone repair products during operation and as a scaffold, and PEGylated PHB has shown an improvement in the intestinal absorption of insulin that was achieved through sustained hypoglycemic outcome beyond 24 hours.

In comparison, chemically synthesized polymers are formed by polylactic acid (PLA), polylactic-co-glycolic acid (PLGA), polyurethane (PU), polyester (methyl methacrylate), polyester (PVP), silicon gum, and polyvinyl alcohol. The biocompatibility and biodegradability of PLA and its copolymer may be obtained from several sources of raw materials. They are classified as renewables, nontoxic, and biodegradable by the Food and Drug Administration. The materials are concrete, mechanically durable, elastic, and thermal materials used as nanocarriers to develop bone tissue, regenerate cartilage, repair cartilage, and formulate controlled-release drugs. The PLA and its copolymers are short-lived, poorly durable, and rapidly degrading in nature, due to its role as carriers in the production of controlled-release medications, the dosage can be decreased more effectively, which eventually enhances the concentration, and side effects in humans, particularly on the liver kidneys, are minimized<sup>9</sup>.

PU polymers have excellent stability (natural, blood, and tissue-compatible), low-stress resistance, wear tolerance, good elasticity, and high strength, compared to other polymer materials. The significant accomplishments of PU include effective clotting and reduced toxicity, nonteratogenic, nonallergic, and noncancerous. As PU applications in various medical biology fields continue to expand, it has the disadvantage that they cannot naturally degrade, ultimately damaging the environment. This is becoming a growing hurdle to PU advancement. The production of biodegradable



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PU materials, therefore, becomes the solution to these problems. Biodegradable PU currently available include PUderived oligosaccharides, lignin, tannin, and PU-derived bark; PU-derived cellulose derivatives; and PU-derived starch.

PLGA has been used in the microsphere, NPs, medical implants, and film preparation of biodegradable polymers with high biocompatibility and efficient degradation. Encapsulated medications in PLGA-NPs or microscopies raise medication dosage, decrease adverse reaction through the aggregation of tumor cells, extend chemotherapy, and enhance pharmaceutical goods' pharmacokinetic impact or microsphere medicines.

#### II. DISCUSSION

Fabrication of functional molecular nanostructures and nanodevices is a cornerstone of many branches of future technologies ranging from electronics, sustainable energy sources, advanced materials, biosensing to medicine. Among many possible strategies, these approaches which are based on site specific, nonlocally induced, on-surface chemical reactions present themselves as exceptionally rewarding. Reactions are triggered on demand and lead to formation of new covalent bonds between molecular precursor units. In this way various 1D and 2D covalently bonded molecular nanostructures, predominantly miscellaneous graphene derivatives, are obtained directly on surface of desired material. In the following article we review current status of research on aryl–aryl coupling on surfaces of semiconducting substrates under ultra-high vacuum conditions. The only successful realizations are reported for rutile titania and hydrogen-passivated germanium surfaces. Due to noticeable differences in chemical nature of these substrates distinct reaction pathways are expected and diverse constraints for efficient on-surface reaction coupling arise.<sup>10</sup>

Supramolecular chemistry deals with the chemistry of the noncovalent bond between molecules and/or ionic species. Inspired by processes in Nature, synthetic receptors mimic biological recognition and regulation processes, and they are applied in catalysis, sensing and separation technologies. Selfassembly of molecular building blocks to larger aggregates forms the basis of nanostructures, molecular materials, and more complex systems. Supramolecular chemistry is the chemical basis for bottom-up nanofabrication.<sup>11</sup>

Heterocyclic compounds possess a cyclic structure with two or more different kinds of atoms in one or more rings. They are widely distributed in Nature and are essential for biological processes. Heterocycles and like pyrimidines and purines carry the genetic information in DNA RNA. Essential amino acids like proline, histidine, and tryptophan are heterocycles that determine the functions of proteins and enzymes. Sugars play important roles in cell-cell recognition and the vast majority of human drugs contain heterocyclic structures. The same holds for pesticides and dyestuffs. The first surface-synthesized 2D covalently-bound molecular nanostructures were reported.<sup>12</sup> The authors demonstrated that the topology of these nanostructures can be engineered by controlling the geometry of the reactive sites on the monomer. They used porphyrin monomers with different numbers and positions of bromine atoms. Two methodologies were adopted to create these nanostructures: the first was to deposit the molecule on Au(111) at room temperature and subsequently anneal the system to cleave the halogen, and the second was achieved by heating the porphyrin monomer in the effusion cell to liberate the bromine atoms, depositing the dehalogenated molecules directly on the surface with no subsequent annealing. To demonstrate the covalent bonding in the structures, Grill et al. manipulated the polymerized molecules using the STM tip to emphasize the robust character of the bonding. In addition, they showed that the periodicity measured between two adjacent polymerized porphyrins is in agreement with DFT simulations.<sup>13</sup>

Using a two-step reaction, Cai et al. demonstrated the fabrication of graphene nanoribbons (GNRs).Experiments were performed using different monomers on the Au(111) and Ag(111) surfaces. The formation of armchair ribbons was achieved by the deposition of monomers onto a surface held at 500 K and 550 K respectively to induce a dehalogenation process and radicals addition. Next, systems 1 and 2 were annealed respectively to 700 K and 740 K, so that cyclo-dehydrogenation took place. To confirm these reactions and exemplify the quality of GNRs, the authors

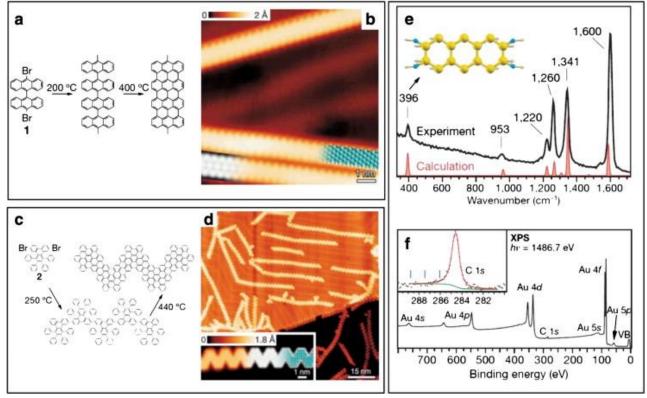


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used additional techniques including XPS and Raman to quantify, respectively, the quality and the width of the GNRs. Thus, as shown in Fig. 3e, the sharp peak at  $396 \text{ cm}^{-1}$  exhibits what is called width-specific radial-breathing-like mode demonstrating experimentally that the radial-breathing-like mode is highly sensitive for probing the GNRs. XPS spectra acquired after the polymerization of monomer 2 indicate only the presence of C1s at 284.5 eV and the presence of other components such as CO, CO or COOH was not observed which means that GNRs are chemically pure and inert under ambient conditions.<sup>14</sup>



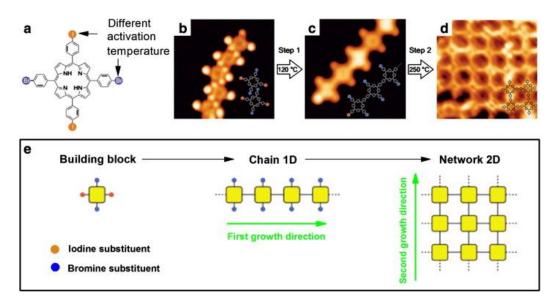
Recently, Grill et al. demonstrated that the use of selective and sequential activation of specific sites on the monomer to facilitate hierarchical 2D polymer growth . A porphyrin molecule with two different types of halogen (bromine and iodine) in the trans configuration was used as monomer building block .The isolated monomer was identified in STM images by its square shape, attributed to the porphyrin block, with two sets of diametrically opposed protrusions of different brightness. The authors attribute the brighter ones to the iodine atoms and the darker to the bromine atoms<sup>15</sup> .To control the sequential activation, Grill et al. deposited the monomer on Au(111) held at 80 K, which preserved the molecules intact on the surface . Heating the system to 420 K led to the selective dissociation of the iodine atoms, while leaving the bromine atoms attached to the monomers, and consequently to the formation of a 1D polymer structure . The formation of 2D polymer networks was subsequently achieved by annealing the system to 550 K . The authors describe this in terms of the dissociation of the bromine atoms from the 1D chains and the subsequent diffusion to form C-C interchain bonds. The mobility of the 1D polymer structure was verified by an STM study.<sup>16</sup> To demonstrate control of the geometry and composition of the covalent system, the authors deposited both trans-Br<sub>2</sub>I<sub>2</sub>TPP and dibromoterfluorene (DBTF) on Au(111) and heated the system up to 550 K. The two-step polymerization reaction was achieved first by forming 1D chains of TPP blocks upon cleaving iodine atoms and secondly by connecting the DBTF monomers to the 1D polymer of TPP after the dissociation of the bromine atoms.<sup>17</sup>



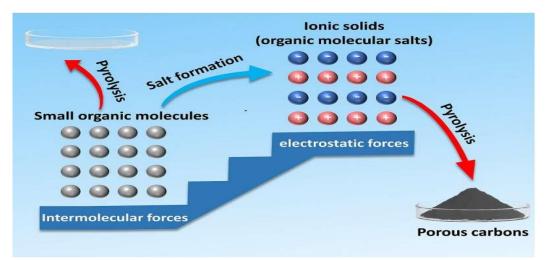
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III. RESULTS



This graphic shows how small organic molecules can be transformed into organic molecular salts, or ionic solids. The organic molecular salts can then be transformed into porous carbons through pyrolysis, which is using high temperatures to activate a transformation. Porous carbons are essential for creating carbon nanostructures, which can be used in a variety of industries.

Small structures made out of carbons are a useful and versatile tool that can be used across industries, including in water and wastewater treatment, gas and oil, and energy storage. In order to create these nanostructures, synthetic and natural polymers have traditionally been used as a starting point to initiate the chemical reaction necessary to create the nanostructured carbons. This is called a precursor.<sup>18</sup>However, both natural and synthetic polymers have limitations. It is difficult to be as precise with natural polymers because of their complexity and synthetic polymers are difficult and expensive to produce.Recent research demonstrated an alternative to polymer precursors by using heat to transform small organic molecules into organic metal salts and then into porous carbons. The process of heating the molecules to



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create a new material is called pyrolysis."The direct purpose of using small molecules as precursors is to simplify carbon preparation by avoiding the polymerization process," said Hai-Wei Liang, a professor and researcher at the University of Science and Technology of China in Hefei."More importantly, this concept of using small molecules can greatly expand the structural diversity of precursors for carbon preparation and thus could pave a pathway to study the relationship between carbon material properties and precursor structures."Previous research into small organic molecules as an alternative to polymer precursors faced limitations due to their synthesis conditions, which created small molecules that were more volatile. This study builds on previous research that showed that using ionic liquids, which is a salt in a liquid state, could resolve some of these limitations. They took this idea one step further and used organic metal salts, also called ionic solids, instead of ionic liquids, because organic metal salts are both organic and inorganic materials. This combination of organic and inorganic materials helps the organic metal salts form templates for the carbon nanostructures. They also show that a wide range of small organic molecules can be used as precursors, as long as they include acidic groups that can transform into salt.<sup>19</sup>

"The difficulty of using small molecules for carbon preparation mainly comes from their high volatility, which can be easily overcome by the transformation of small molecules into organic metal salts. This is because the original weak intermolecular force holding molecules together is replaced by robust electrostatic force after the salt formation, thus lowering the volatility," said Liang. Another benefit of using this method is the versatility of the organic metal salts. By altering the components of the organic metal salts, the characteristics of the carbon nanostructures can be controlled at a molecular level. Looking ahead, researchers will continue to explore the different ways this technique can be used. "Next, we will continue to explore the structural relationship between carbon materials and molecular precursors to establish well-defined rules to guide the rational synthesis of carbon materials at the molecular level. Ultimately, we hope to use the advantage of this method in controlling carbon structures and compositions to achieve the tailor-made synthesis of advanced functional carbon materials for targeted applications," said Liang.<sup>20</sup>

#### **IV. CONCLUSIONS**

Recent results on synthesis, structural, and physicochemical studies of complexes based on detonation nanodiamonds modified with metal-organic molecules with functional magnetic and fluorescent properties have been presented. Neutron scattering experiments have discovered subtle features of the assembly of electro-negative molecules of Eu diphthalocyanines and nanodiamonds with positive surface potential in aqueous media. Nanoscale ordering of diamonds reinforced by their linking via diphthalocyanines was confirmed by TEM. The stable complexes possess Xray luminescent and magnetic properties.<sup>21</sup> Such nanostructures with low toxicity are prospective as contrasting and photoactive agents for magnetic resonance imaging and photodynamic therapy. The design of hybrid nanostructures extends a variety of biological materials with antibacterial activity. This is actually due to bacterial resistivity to widely used antibiotics. For example, in-situ growth of Ag particles on cellulose nanocrystals allowed create the scaffolds with robust antibacterial activity against E. coli.For other purposes (bone regeneration, cell growth stimulation), there was realized electrostatic self-assembly of positively charged magnetite nanoparticles with electro-negative graphene oxide, and these complexes were incorporated into poly L-lactic acid scaffolds. <sup>22</sup>Special attention was paid to the hybrids based on polymers and inorganic particles, shown the improvements in drug targeting comparative to non-hybrid platforms. Various hybrids serve for gene therapy and phototherapy, tissue regeneration, vaccines, relevant applications in theranostics. Among hybrid systems with different particles (polymeric, metal, oxide, and carbon),<sup>26</sup> the diamondbased platforms for drug delivery and bioimaging are the most prospective structures due to the unique chemical and physical properties of diamonds (chemically inert, resistant to irradiation, luminescent, etc.). Since diamonds combine both types of carbon orbital hybridization, it provides them also with photo- and electro-catalytic properties important for biology. Moreover, small particles of detonation nanodiamonds (DNDs) <sup>24</sup>are well ordered in aqueous dispersions.<sup>1</sup> This makes them real candidates to be antiviral platforms like the shells capturing viruses to decrease (inhibit) infection. To create such platforms, one needs to regulate the aggregation of diamonds via surface potential by grafting surface groups (COOH, OH, H) and by modification with biomolecules. The valuable applications of



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diamonds can be provided by grafting metal ions (magnetic and luminescent) to their surface to use them in magnetic resonance imaging (MRI) and photodynamic therapy with X-rays (X-PDT). <sup>25</sup> In the latter case, due to optical emission, it is generated desirable singlet oxygen chemically active in biological tissues. The alternative way is to use the DNDs modified with the molecules carrying magnetic atoms (4 f, 3d) having luminescent properties needed to design new agents for MRI and X-PDT.<sup>23</sup>

We developed the method of DNDs association with Eu diphthalocyanines (DPC) resistive to thermal, chemical, and radiation treatment and keeping firmly metal atoms. The first prepared DPC-DND complexes examined by physicochemical, X-ray fluorescence methods were studied by small-angle neutron scattering (SANS) in solutions comparatively to pure diamond dispersions, examined by TEM and tested on living cells to estimate the toxicity of new substances.<sup>26</sup>

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