Functional Nanowires

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Abstract

Nanotechnology offers the promise of enabling revolutionary advances in diverse areas ranging from electronics, optoelectronics, and energy to healthcare. Underpinning the realization of such advances are the nanoscale materials and corresponding nanodevices central to these application areas. Semiconductor nanowires and nanobelts are emerging as one of the most powerful and diverse classes of functional nanomaterials that are having an impact on science and technology. In this issue of *MRS Bulletin*, several leaders in this vibrant field of research present brief reviews that highlight key aspects of the underlying materials science of nanowires, basic device functions achievable with these materials, and developing applications in electronics and at the interface with biology. This article introduces the controlled synthesis, patterned and designed self-assembly, and unique applications of nanowires in nanoelectronics, nano-optoelectronics, nanobiotechnology, and energy harvesting.

Nanowires As Building Blocks for Bottom-Up Nanotechnology

The field of nanotechnology represents an exciting and rapidly expanding research area that crosses the borders between the physical, life, and engineering sciences.¹ Much of the excitement in this area of research has arisen from recognition that new phenomena, multifunctionality, and unprecedented integration density are possible with nanometer-scale structures. In general, there are two philosophically distinct approaches for creating small objects: top-down and bottom-up.

In the top-down approach, small features are patterned in bulk materials by a combination of lithography, etching, and deposition to form functional devices and their integrated systems. The top-down approach has been exceedingly successful in many venues, with microelectronics being perhaps the best example today. While developments continue to push the resolution limits of the top-down approach, these improvements in resolution are associated with a near-exponential increase in cost associated with each new level of manufacturing facility. This economic limitation and other scientific challenges with the top-down approach, such as making nanostructures with near-atomic perfection and incorporating materials with distinct chemical and functional properties, have motivated efforts worldwide to search for new strategies to meet the demand for nanoscale structures today and in the future.^{2–4}

The bottom-up approach, in which functional structures are assembled from well-defined chemically and/or physically synthesized nanoscale building blocks, much like the way nature uses proteins and other macromolecules to construct complex biological systems, represents a powerful alternative approach to conventional top-down methods.3,5 The bottom-up approach has the potential to go far beyond the limits and functionality of top-down technology by defining key nanometer-scale metrics through synthesis and subsequent assembly-not by lithography. Moreover, it is highly likely that the bottom-up approach will enable entirely new device concepts and functional systems and thereby create technologies that we have not yet imagined. For example, it is possible to seamlessly combine chemically distinct nanoscale building blocks that could not be integrated together in top-down processing and thus obtain unique function and/or combinations of function in an integrated nanosystem.

To enable this bottom-up approach for nanotechnology requires a focus on three key areas that are at the heart of devices and integration. First, the bottom-up approach necessitates nanoscale building blocks with precisely controlled and tunable chemical composition, structure, size, and morphology, since these characteristics determine

their corresponding physical properties. To meet this goal requires developing methods that enable rational design and predictable synthesis of building blocks. Second, it is critical to develop and explore the limits of functional devices based on these building blocks. Nanoscale structures may behave in ways similar to current electronic and optoelectronic devices, although it is also expected that new and potentially revolutionary concepts will emerge from these building blocks, for example, due to quantum properties. Third and central to the bottom-up concept will be the development of architectures that enable high-density integration with predictable function, and the development of hierarchical assembly methods that can organize building blocks into these architectures.

Nanowires (NWs)3,6 and nanobelts (NBs)^{7,8} represent an important and broad class of one-dimensional (1D) nanostructures at the forefront of nanoscience and nanotechnology. NWs and NBs are typically single-crystalline, highly anisotropic, semiconducting, insulating, and/or metallic nanostructures that result from rapid growth along one direction. The cross section of NWs and NBs is uniform and much smaller than the length. NWs are typically cylindrical, hexagonal, square, or triangular in cross section; NBs are typically rectangular in cross section, with a large anisotropy in dimensions. NWs and NBs can be rationally and predictably synthesized in single-crystal form with all key parameters controlled during growth, including chemical composition, diameter, length, doping, growth direction, and possibly surfaces.^{9,10} NWs thus represent one of bestdefined and controlled classes of nanoscale building blocks compared to, for example, carbon nanotubes. The unique control over the microstructure of NW building blocks arises from an excellent understanding of their growth mechanisms and the broad range of chemical compositions achievable (versus simply carbon).^{6–14} Such control has enabled a wide range of devices and integration strategies to be pursued in a rational manner. For example, semiconductor NWs have been assembled into nanometer-scale field-effect transistors (FETs),^{11–13} p-n diodes,^{12,14} light-emitting diodes (LEDs),12 bipolar junction transistors,¹⁴ complementary inverters,¹⁴ nanoscale lasers,¹⁵ complex logic gates, and even computational circuits that have been used as basic digital calculators,¹⁶ gas sensors,¹⁷ nanoresonators,^{18,19} and nano-generators.²⁰ Polar-surface-dominated NBs spontaneously self-assemble into nanosprings,^{21,22} nanobows,²³ nanorings,²⁴ and nanohelixes.²⁵ which are candidates for nanoscale transducers, actuators, and

sensors.^{26,27} These advances and the growing interest in NWs and NBs can be quantified in terms of the rapidly increasing number of publications per year since the early 1990s (Figure 1).

Nanowires and Carbon Nanotubes

Another class of 1D nanostructures that have received considerable attention is carbon nanotubes (NTs).28 In contrast to NTs whose electronic properties are largely determined by the chirality of the graphene layers, NWs offer several unique merits. First, NW devices can be assembled in a rational and predictable manner because the size, interfacial properties, and electronic properties of the NWs can be precisely controlled during synthesis. Moreover, reliable methods exist for their parallel assembly.²⁹ The growth direction and side surfaces of nanowires can be precisely controlled during synthesis to produce desired properties. Second, it is possible to combine distinct NW building blocks in ways not possible in conventional electronics and to leverage the knowledge base that exists for the chemical modification of inorganic surfaces30,31 to produce semiconductor NW devices that achieve new function and correspondingly could lead to unexpected device concepts. Finally, the structure of NWs can be rationally designed both axially and radially for integrating materials of different chemical composition and possibly crystal structure into an entity that exhibits multifunctionality and integrated properties at the nanoscale. Therefore, NWs open up a new paradigm in

nanotechnology well beyond that possible with a single-composition building block.

Designed Synthesis of Nanowires and Nanowire Heterostructures

The rational design and synthesis of nanoscale materials is critical to work directed toward understanding fundamental properties, creating nanostructured materials, and developing nanotechnology. Strategies have been developed to design and rationally synthesize NWs and NBs with predictable control over the key structural, chemical, and physical properties. Among the numerous methods that have been explored, a strategy that has received increasing focus in the past several years involves exploiting a "catalyst" to confine growth in 1D. Depending on the phases involved in the reaction, this approach is typically defined as vapor-liquid-solid (VLS),32 solution-liquid-solid (SLS),^{33,34} or vapor-solid (VS)³⁵ growth. In VLS growth, the catalyst is envisioned as a nanodroplet that defines the diameter of and serves as the site that preferentially directs the addition of reactant to the end of a growing NW, as indicated in Figure 2a by the yellow region at the left-hand end of the NW.

Modulated nanostructures in which the composition and/or doping are varied on the nanometer scale represent important targets of synthesis since they could enable new and unique function and potential for integration in functional nanosystems. The approach to axial NW heterostructure growth (Figure 2b)³⁶ exploits metal-catalyzed nanowire synthesis. To create a



Figure 1. Plot illustrating the number of published papers relating to nanowires each year since the early 1990s. In the background is a scanning electron microscopy image of fine silicon nanowires (average diameter, 20 nm).

single junction within the NW, the addition of the first reactant is stopped during growth, and a second reactant is introduced for the remainder of the synthesis; repeated modulation of the reactants during growth produces NW superlattices. In principle, this approach can be successfully implemented if a nanocluster catalyst suitable for growth of the different superlattice components under similar conditions is found. Gold nanoclusters meet this requirement for a wide range of Group III-V and Group IV materials. This methodology for the growth of superlattice structures can be generalized in many materials systems. Structures have been fabricated for p-n junctions within individual Si NWs by Au-nanoclustercatalyzed chemical vapor deposition (CVD) and dopant modulation.36 Similar structures have also been demonstrated for Si-Ge³⁷ and InAs-InP.³⁸ These superlattice structures greatly increase the versatility and power of NW building blocks for nanoscale electronic and photonic applications such as nanobarcodes, injection lasers, and engineered 1D waveguides.

Designed Synthesis of Radial Nanowire Homo- and Heterostructures

The growth of crystalline overlayers on nanostructure surfaces is important for controlling surface properties and enabling new function. This concept has been proved by the synthesis of silicon and germanium coreshell and multishell NW homo- and heterostructures using the CVD method applicable to a variety of nanoscale materials.39 Axial growth is achieved when reactant activation and addition occurs at the catalyst site and not on the NW surface through epitaxial growth (Figure 2b). Correspondingly, it is possible to drive conformal shell growth by altering conditions to favor homogeneous vapor-phase deposition on the NW surface. Subsequent introduction of different reactants and/or dopants produces multishell structures of designed composition, although epitaxial growth of these shells requires consideration of lattice structures.

Designed Synthesis of Hierarchical Structured Nanowire Networks

In VLS growth, the location at which the NW grows is defined by the site of the catalyst particle, and the orientation of the NW is determined by the surface lattice of the substrate on which an epitaxial relationship can be built. Based on such a principle, by decorating a grown radial/axial NW heterostructure with catalyst particles, side branches can be grown along the orientations defined by the least lattice



Figure 2. Schematic illustration of the evolution of nanowire structural and compositional complexity enabled today by controlled synthesis, from (a) homogeneous materials to (b) axial and radial heterostructures and (c) branched heterostructures. The colors indicate regions with distinct chemical composition and/or doping.

mismatch between the NW "substrate" and the newly grown branches, forming 3D networks⁴⁰ (see the articles by Bakkers et al. and Dick et al. in this issue). The materials for the main branches and the side branches can be of different chemical compositions, resulting in the formation of 3D junction arrays (Figure 2c). Growth of III–V NWs on Group IV substrates enables the integration of high-performance III–V semiconductors monolithically with silicon technology, since fundamental issues of III–V integration on Si, such as lattice and thermal expansion mismatch, can be overcome.

Nanowires of Functional and Smart Materials

Complex oxides with structures such as perovskite, spinel, and garnet have many important properties and applications in science and engineering including ferroelectricity, ferromagnetism, colossal magnetoresistance, luminance, optoelectronics, and semiconductors. These properties are crucial for applications such as data storage, data retrieval, and sensing. Besides the synthesis of conventional binary oxides (discussed by J. Wang et al. in this issue), methods have been developed to synthesize NWs of functional and smart materials. Liu et al.⁴¹ have demonstrated a generic approach for the synthesis of single-crystal complex oxide nanostructures of perovskites, spinels, and various hydroxides, including monoclinic, corundum, CaF2structured, tetragonal, and metal hydroxides.42,43 The method is based on a reaction between a metallic salt and a metallic oxide in a solution of composite hydroxide eutectic at $\sim 200^{\circ}$ C and 1 atm without using an organic dispersant or capping agent. This one-step synthesis technique is cost-effective, easy to control, and can be conducted at low temperature and normal atmospheric pressure. The technique can be expanded to many materials systems, and it provides a general, simple, convenient, and innovative strategy

for synthesizing nanostructures of complex oxides with important scientific and technological applications in ferroelectricity, ferromagnetism, colossal magnetoresistance, fuel cells, optics, and more.

Organization and Assembly of Nanowires

The organization and/or directed assembly of NWs into designed hierarchical structures is a key goal necessary for creating arrays of devices. Using a patterned catalyst, NWs can be directly grown on a solid substrate in a designed configuration. This strategy enables nanoscale structures to be produced directly on a 2D substrate without extensive lithography, but it still faces many of the traditional constraints of planar growth and device fabrication.

Alternatively, NW materials produced under synthetic conditions optimized for their growth can be organized into arrays by several techniques including (1) electricfield-directed, (2) fluidic-flow-directed, (3) Langmuir–Blodgett, and (4) patterned chemical assembly.

Applied electric fields (E-fields) can be used effectively to attract and align NWs due to their highly anisotropic structures and large polarization.¹¹ E-field–directed assembly not only can organize NWs along the electric field, but also can be used to position individual NWs at specific positions with controlled directionality.⁴⁴

Another powerful approach called fluidic-flow-directed assembly¹⁶ allows alignment of NWs (or NTs) by passing a suspension of NWs through microfluidic channel structures. Fluidic-flow-directed assembly can also be used to organize NWs into more complex crossed structures, which are critical for building dense nanodevice arrays, using a layer-by-layer deposition process. The microfluidic method enables the alignment of NWs up to about the millimeter scale.

Assembly on larger scales with precise spacing down to the nanometer level re-

quires alternative strategies. The Langmuir-Blodgett (LB) technique, in which an ordered monolayer is formed on water and transferred to a substrate, represents an alternative method that is beginning to achieve this goal. For example, parallel and crossed NW structures have been assembled by single and sequential transfers over centimeter length scales; moreover, these large-area arrays have been efficiently patterned into repeating arrays of controlled dimensions and pitch to yield hierarchical structures with order defined from the nanometer through the centimeter length scales.45 In addition, patterned chemical modification of a substrate can be an effective approach for directedor self-assembly of NWs and NTs.^{16,45}

Nanoelectronic Devices

Homogeneous doped NWs represent key building blocks for a variety of electronic devices.^{16,46–51} A prototypical example of such a device with broad potential for applications is the NW field-effect transistor (NWFET). For example, studies of NWFETs fabricated from boron-⁴⁶ and phosphorus-doped⁴⁷ Si NWs have shown that the devices can exhibit performance comparable to the best reported for planar devices made from the same materials. Studies have also demonstrated the high electron mobility of epitaxial InAs NWFETs with a wrap-around cylindrical gate structure surrounding a nanowire.⁴⁸

More generally, controlled bottom-up assembly and synthetic elaboration of NWs offers unique opportunities. The crossed NW architecture enables device properties to be defined by the assembly of the NW components and not by lithography, and has been utilized to demonstrate logic gate structures, basic computation, and selective addressing.16,49 Synthesis of axial modulation-doped NW heterostructures has enabled the creation of address decoders and coupled quantum structures without a critical use of lithography,⁵⁰ while the design of radial Ge/Si core-shell NW heterostructures demonstrated a true performance benefit of NWFETs compared with state-of-the-art planar devices.⁵¹

Nanowire Nanosensors

Field-effect transistors fabricated using individual NWs are ultrasensitive nanosensors for detecting a wide range of gases, chemicals, and biomedical species in both commercial and research applications.^{52,53} The high-performance characteristics of NWFETs, such as high surface-to-volume ratio and specially designed surface structures, are key factors that lead to very high sensitivity. More important to overcoming the sensitivity limitations of previous planar FET sensors is the 1D morphology of these nanoscale structures. Specifically, binding to the surface of a nanowire leads to depletion or accumulation of carriers in the "bulk" of the nanometer-diameter structure versus only the surface region of a planar device.52 NWFETs can be configured as highly selective and highly sensitive detectors by linking recognition or receptor groups to the surface. This was first demonstrated with Si NWFETs, which were used for detection of pH, metal ions, and proteins.54 More generally, these unique features of semiconductor nanowires (see article by Patolsky et al. in this issue) have led to sufficient sensitivity and selectivity to enable the detection of viruses at the single-virus level⁵² and sequence-specific DNA detection at the femtomolar level.53

High-Performance Macroelectronics and Flexible Electronics

A new concept of macroelectronics using assembled semiconductor NW thin films promises significant performance improvement for macroscale electronics. In this approach, a "thin film" of oriented semiconductor NWs aligned by self-assembly techniques is used to produce thin-film transistors (TFTs) with conducting channels formed by multiple parallel singlecrystal NWs (see the article by Duan in this issue). Charges are transported from source to drain within single crystals, ensuring high carrier mobility. Such devices can be made using semiconductor NWs on a wide range of substrates, including polymer and glass, providing a new approach for flexible electronics. The device performance of these NW TFTs not only greatly surpasses that of solution-processed organic TFTs, but is also significantly better than that of conventional amorphous Si TFTs, and is approaching the performance of single-crystal silicon-based devices. Furthermore, Group III-V or II-VI nanowire or nanobelt materials of high intrinsic carrier mobility or optical functionality can be assembled into thin films on flexible substrates to enable new multifunctional electronics and optoelectronics that are not possible with traditional macroelectronics, in which electronic components are distributed over substrates with surface areas on the order of square meters. This can have an impact on a broad range of existing applications, from flat-panel displays to image sensor arrays, and enable a whole new generation of flexible, wearable, or disposable electronics for computing, storage, and wireless communication.

Nanophotonics

Nanowires represent attractive building blocks for active nanophotonic devices, including light-emitting diodes (LEDs), lasers, and detectors.^{11,55-59} Significantly, the ability to assemble and electrically drive nanoscale sources and detector blocks could allow for fully integrated nanophotonic systems for use in applications ranging from biodetection through information processing. The crossed NW approach was the first to demonstrate true nanoscale LEDs, or nanoLEDs (Figure 3a). In this work, nanoscale p-n diodes were created by crossing well-defined p-type and *n*-type InP NWs, and subsequent device measurements showed that bandedge emission is observed at the nanoscale cross-points in forward bias.¹¹ This concept has enabled the assembly of a wide-range of nanoLEDs on a single chip, with emission ranging from ultraviolet through near-infrared in a manner not possible with conventional planar technology.55 The crossed NW architecture can be further generalized to hybrid devices consisting of *n*-type direct-bandgap NWs assembled onto p-Si electrodes defined in heavily *p*-doped planar substrates. Significantly, using *n*-type CdS NWs in this type of nanostructure has led to the demonstration of the first nanoscale electronic injection laser (Figure 3b).56 In addition to nanoscale light sources, crossed NW p-njunctions can also be configured as photodetectors critical for integrated photonics. For example, avalanche multiplication of the photocurrent in nanoscale p-n diodes



Figure 3. Nanowire photonics. (a) Nano-LED based on the crossed nanowire approach. The upper image corresponds to an electroluminescence image recorded from the device shown in the lower AFM image. The peak emission occurs at the cross-point of the two nanowires. (b) Nanoscale electronic injection laser using n-type CdS nanowires. The upper optical image shows the position of the nanowire end on right side and upper rectangular electrode, while the lower electroluminescence image shows light emission only from the nanowire end of the laser. (c) Current/voltage plot of a crossed Si-CdS nanowire p–n diode as a nanoscale photon detector. The black curve shows the background signal, while the red curve shows the signal in the presence of light. The inset shows an optical image of a two-element crossed nanowire (vertical); the larger rectangular features correspond to metal contacts. (d) The nanoscale photon detector arrays can be addressed independently without electrical crosstalk (1_{oh} is photocurrent).

has recently been shown in crossed Si-CdS NWs (Figure 3c).⁵⁷ These NW avalanche photodiodes, or nanoAPDs, exhibit ultrahigh sensitivity with detection limits of less than 100 photons and subwavelength spatial resolution of 250 nm. Moreover, the elements in nanoAPD arrays can be addressed independently without electrical crosstalk. These characteristics exceed the capabilities of any known detector and could open up unique opportunities, for example, for integrated lab-on-a-chip devices and imaging of biological systems.

The controlled synthesis of radial NW structures also offers substantial opportunities for nanophotonics, since the required *n*- and *p*-type active materials can be incorporated directly as the core and shells in the NW. This general approach was first demonstrated with the growth of well-defined doped III-nitride–based core–multishell NW heterostructures (Figure 4a).^{58,59} In these nanostructures, an *n*type GaN core and *p*-type GaN outer shell serve as electron and hole injection layers, and an In_xGa_{1-x}N shell provides a tunablebandgap quantum well for efficient radiative recombination of injected carriers.

Nanophotonic devices with separate contacts to the *n*-type core and *p*-type outer shell show expected p-n diode current rectification (Figure 4b). In forward bias, the devices yield strong light emission with the LED color dependent on the indium composition. Significantly, nano-LED spectra collected from such radial NW heterostructure devices have shown a systematic redshift from 367 nm to 577 nm (Figure 4c), covering the shortwavelength region of the visible spectrum, and moreover, they indicate that very high quantum efficiencies will be possible in such defect-free structures. The efficient injection and radiative recombination of carriers, as well as the synthetically tunable emission wavelength of radial NW devices, represent a clear advance in nanoLED sources and thus a promising pathway to multicolor NW injection lasers in the future.

Nano-Biotechnology

Integration of nanosystems and biosystems is a multidisciplinary field that has the potential for tremendous impact on biology, chemistry, physics, biotechnology, and medicine. The combination of these diverse areas of research promises to yield revolutionary advances in healthcare, medicine, and the life sciences through, for example, the creation of new and powerful tools that enable direct, sensitive, and rapid analysis of biological and chemical species. Patolsky et al.⁵² have demonstrated the first application of NW nanosensors for ultrasensitive



Figure 4. Radial nanowire core-shell heterostructures as nanophotonic building blocks. (a) Schematic of core-multishell nanowire with n-GaN/ InGaN/GaN/ p-AIGaN/ p-GaN structure. GaN is blue, InGaN is green, and p-AlGaN is yellow. (b) Current-voltage data recorded from a device with contacts on the n-core and p-shell. Inset is a scanning electron micrograph of the fabricated device using the synthesized structure. (c) Normalized electroluminescence spectra recorded from five multicolor core-shell nanowire LEDs with (left to right) 1%, 10%, 20%, 25%, and 35% indium, respectively.

detection of proteins down to individual virus particles as well as multiplexed recording of these species using distinct NW elements within a sensor device. In addition, Patolsky et al.⁶⁰ have demonstrated an unprecedented approach for investigating the electrical properties of hybrid structures consisting of arrays of NWFETs integrated with the individual axons and dendrites of live mammalian neurons, where each nanoscale junction

can be used for spatially resolved, highly sensitive detection, stimulation, and/or inhibition of neuronal signal propagation. Details are described by Patolsky et al. in this issue. Arrays of nanowire-neuron junctions enable simultaneous measurement of the rate, amplitude, and shape of signals propagating along individual axons and dendrites. The configuration of nanowire-axon junctions in arrays, as both inputs and outputs, makes possible controlled studies of partial to complete inhibition of signal propagation by both local electrical and chemical stimuli. This revolutionary development opens a new field in integrated nano-biotechnology.

Nanoelectromechanical Systems

The development of novel technologies for wireless nanodevices and nanosystems is critically important for *in situ*, real-time, and implantable biosensing and biomedical monitoring. Nanosensors are currently under intense development for ultrasensitive and real-time detection of biomolecules.

An implanted wireless biosensor, for example, requires a power source, which may be provided directly or indirectly. It is highly desirable for wireless devices (and required for implanted biomedical devices) to be self-powered without the need for finite-lifetime batteries. Using aligned ZnO NWs grown either on a crystal substrate or a polymer substrate,⁶¹ an innovative approach has been demonstrated for converting nanoscale mechanical energy into electric energy.²⁰ By deflecting the aligned NWs using a conductive atomic force microscope (AFM) tip in contact mode, the energy that was first created by the deflection force and later converted into electricity by the piezoelectric effect has been measured to demonstrate a nanoscale power generator. The operation mechanism of the electric generator relies on the unique coupling of piezoelectric and semiconducting dual properties of ZnO as well as the elegant rectifying function of the Schottky barrier formed between the metal tip and the NW.⁶²

This research demonstrates the feasibility of harvesting energy from the environment, such as converting mechanical energy (e.g., body motion or muscle stretching), vibrational energy (e.g., acoustic or ultrasonic waves), and hydraulic energy (e.g., body fluid and blood flow) into electric energy for self-powered nanosensors and nanosystems. It also has a huge impact on miniaturizing the size of integrated nanosystems by reducing the size of the power source and improving its efficiency and power density. Piezoelectric FETs and diodes as well as force sensors have also been demonstrated using NWs.

Conclusions

The development of nanowire- and nanobelt-based materials represents breakthrough achievements with rapid expanding impact in all areas of nanotechnology. The remarkable level of synthetic control of the performance properties of NWs and NBs is leading to revolutionary technologies in electronics, optoelectronics, sensors, the life sciences, and defense and will continue to broadly impact the fields of physics, chemistry, biology, medicine, environmental science, and engineering. The articles in this issue of MRS Bulletin make it clear that NWs are truly powerful building blocks for achieving the bottom-up paradigm of nanotechnology with demonstrated impacts on both fundamental science at the nanoscale and on applications. Looking forward, we believe that the future is remarkably bright, with likely revolutionary technologies from NW-based nanosystems that will impact in many ways such areas as the life sciences, healthcare, information technology, and energy science, to name just a few.

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