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REVIEW

Clean Water through Nanotechnology: Needs, Gaps, and Fulfillment

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ABSTRACT: Sustainable nanotechnology has made substantial contributions in providing contaminant-free water to humanity. In this Review, we present the compelling need for providing access to clean water through nanotechnology-enabled solutions and the large disparities in ensuring their implementation. We also discuss the current nanotechnology frontiers in diverse areas of the clean water space with an emphasis on applications in the field and provide suggestions for future research. Extending the vision of sustainable and affordable clean water to environment in general, we note that cities can live and breathe well by adopting such technologies. By understanding the global environmental challenges and exploring remedies from emerging nanotechnologies, sustainability in clean water can be realized. We suggest specific pointers and quantify the impact of such technologies.

KEYWORDS: *clean water, nanotechnology, desalination, atmospheric water harvesting, nanosensors, toxicity, smart water purifiers, Internet of Things*

Crowded, expanding cities in many parts of the world are experiencing an increased demand for fresh water, and planners are unclear as to how the water needs of tomorrow will be met. In cities such as Bangalore, where data are currently the most valuable commodity, we believe that a data ecosystem could be created for water. Focus on water availability is likely to create businesses, drive the economy, and make the world breathe better. Taking a specific case, India has just 4% of the global freshwater resources but ∼18% of the world's population. The country, which was largely rural years ago, has *en masse* become urban in the past two decades. The urban population has risen from 28% in 2000 to 33% in 2016.[1] With a growth rate over 6% in gross domestic product (GDP), the most populous countries, such as India and China, are increasing their chemical, pharmaceutical, agrochemical, automotive, petrochemical, semiconductor, and many other outputs, which will eventually "enrich" our ecosystem materially. Simultaneously, their rapidly declining water resources will be burdened by unprocessed industrial waste. The World Bank has predicted that achieving a growth rate of 8% or above for India will be possible only with a robust water management system.[2] These emerging issues, similar to those existing throughout the world, present a complicated suite of problems that will require technological advances, limits on usage, and collective wisdom, and compassion in order to create sustainable solutions. For instance, the control over carbon emissions by developed countries is probably not the reason for the globe's survival, but the lack of development in less-developed countries is, according to the Intergovernmental Panel on Climate Change (IPCC).[3] Sustainable economic and technological development for all is needed, although acquiring a quality of life comparable to the United States for the rest of the world would require significant advances in treating, purifying, and assessing toxicity in water. Clean water challenges are highly interdisciplinary, and solutions therefore must cut across boundaries of disciplines. Water in diverse forms is related to climate, food, health, and many other aspects of life, including its origin. The need for urgent, concerted action is clear from just one observation: *ca.* 83% of freshwater species have declined in the last 50 years.[4] Water is and will continue to be one of the most important interdisciplinary subjects of research.

Recent advances in the field of nanoscience provide many solutions to alleviate needs with regard to reducing scarcity or removing contamination. For example, there are filters that remove pesticides from drinking water using nanochemis-

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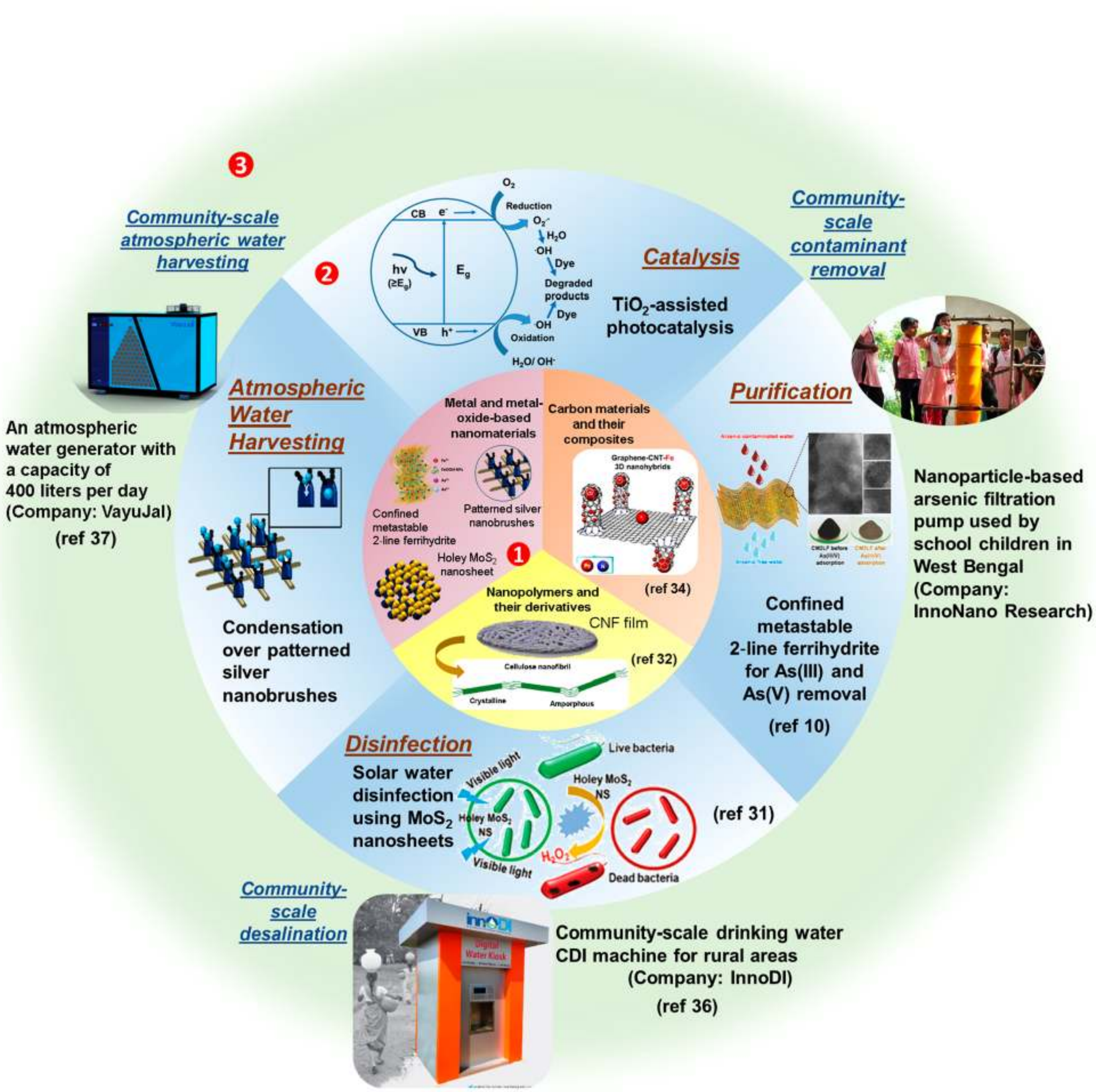

Figure 1. Schematic illustrating translation of materials from lab-scale to market. Innermost circle 1 indicates materials as building blocks; middle circle 2 indicates reported phenomena using such materials; and outermost circle 3 shows products built out of research and their commercialization to create a societal impact. Images containing 2-line ferrihydrite in circles 1 and 2 adapted with permission from ref 10. Copyright 2017 John Wiley and Sons. Images involving holey MoS$_2$ in circles 1 and 2 adapted with permission from ref 31. Copyright 2018 John Wiley and Sons. Image in yellow section of circle 1 adapted with permission from ref 32 under Creative Commmons Attribution 4.0 International License. Schematic showing graphene-CNT-Fe nanohybrids in circle 1 adapted with permission from ref 34. Copyright 2013 American Chemical Society. Image of a community-scale CDI machine in circle 3 adapted with permission from ref 36. Copyright 2018 Innodi Water Technologies Pvt. Ltd. Image of an atmospheric water generator in circle 3 adapted with permission from ref 37. Copyright 2019 VayuJal Technologies Pvt. Ltd.

try.[5−8] This technology had already reached over 7.5 million people by 2016, when implementation data were last collected, reducing pesticide levels from over 20 times the safety standard to concentrations substantially below it (0.5 parts per billion, ppb, for all pesticides taken together).[9] In another example, a nanostructured material is able to remove arsenic from drinking water affordably and the technology is delivering clean water (CW) to about 1 million people each day, providing hope for another 80 million or so in India, who are affected by this problem.[10−12] The government of India has approved the technology for national implementation. Such a solution does not require electricity and is affordable, even for those living in the poorest parts of the world. Several alternate solutions to address arsenic as well as other organic and inorganic contaminants are available and are being explored in various parts of the world. Alternate methods of microbial disinfection, desalination, water harvesting, recycling, contaminant sensing, and monitoring are debuting in the marketplace. Scalability and massive implementation of technologies is slow but encouraging. For example, the prospects of nanotechnology (NT) for CW have enthused many researchers, and numerous articles have been published around the theme of NT and nanomaterials for CW production and wastewater treatment.[13,14] Among several issues of relevance for developing affordable NTs for CW, there are four principal points to be considered: (1) More for less: As constituent materials reduce in dimension and reach the nanoscale regime, their effective capacity to remove contaminants increases due to additional derivatization of the material to increase charge, solubility, affinity, *etc*. Properties of relevance, such as the presence of active surface sites for adsorption, enhanced adsorption enthalpy for specific species, reactivity as a result of activation of specific chemical bonds, size-dependent optical absorption, and emission, are being explored today. All of these properties individually and collectively make it possible to acquire more effective scavenging capacity per unit mass of the material at the nanoscale than the bulk material, making a purifier composed of nanoscale material smaller and more affordable. (2) Decreasing limits of contaminants: The World Health Organization (WHO) set the upper limit on arsenic in drinking water at 50 ppb in 1963,[15] and the U.S. Environmental Protection Agency decreased the limit to 10 ppb in 2002.[16] A primary reason for decreasing contaminant limits has been an enhanced understanding of the effects of contaminants on human health. However, the limited availability and high cost of remedial technologies have kept governments from implementing these standards. Moreover, the actual safe limit is expected to be further below the present WHO limit. This is because the arsenic intake per capita per day through drinking water is much higher than that assumed by the WHO in the arsenic-affected tropical regions of India due to unaccounted sources of arsenic intake, such as food crops.[17] In addition to India, South Asian countries including Vietnam and Bangladesh,[18−21] and Latin American countries including Argentina, Bolivia, Chile, and others including regions of the United States are also prone to alarming levels of arsenic.[21−23] In order to accomplish the already prescribed limits, nanomaterials are essential. (3) What are achievable levels of contaminant removal using advanced materials? Single nanoparticles (NPs) have even shown sensitivity to a few species of contaminants.[24] Thus, even at this level of contaminant concentration, several materials are selective in removing them. (4) Special properties: The unique properties of nanostructures, such as atomically precise pores and thicknesses of desired dimensions,[25] controlled functionalities,[26] maneuverability,[27] *etc.*, offer exciting possibilities for making CW.

In this Review, we discuss nanomaterials and technologies that can be used for water treatment and sensing, highlighting major challenges that need to be addressed through NT in providing sustainable access to CW.

NANOMATERIALS AND NANOTECHNOLOGIES THROUGH THE AGES

Throughout history and into the present, water filtration and purification components are primarily made of carbon. Using carbon in the form of wood charcoal for water purification was practiced by Egyptians and Sumerians in 3750 BC.[28] Activated carbon, introduced in the 1940s,[29] and its various modifications, including nanostructures such as carbon nanotubes (CNTs), carbon nanofibers (CNFs), and graphene-based materials have been exploited for treatment through mechanisms such as adsorption, catalytic wet air oxidation, membrane-based separation, and disinfection, and also for sensing and monitoring.[30] Metal and metal oxide NPs have also been reported for adsorption, photocatalysis, oxidation, disinfection, and sensing. Despite being an active research area for decades, nanomaterials-based treatment and sensing technologies are yet to occupy a large share of the market due to unreliability in terms of sensitivity and selectivity, higher cost, and field-level issues during operation. Figure 1 illustrates applications of nanomaterials, such as, MoS_2 nanosheets for disinfection,[31] CNF films for biocidal activity,[32] silver nanobrushes for atmospheric water harvesting,[33] graphene-CNT-iron oxide nanostructures,[34] and ferrihydrite for heavy-metal removal.[10] These have reached people in the form of affordable and easy-to-operate devices such as filtration-incorporated hand pumps,[35] desalination units,[36] and atmospheric water generators.[37] We discuss the future of nanomaterials that can be integrated into these technologies to overcome the existing challenges.

DESALINATION

Nearly 40% of the global population resides within 100 km of an ocean or a sea, rendering desalination a crucial solution to water scarcity. Presently, there are 19,744 desalination plants operating across 150 countries supplying 100 million m^3 of water per day to 300 million people globally.[38] However, desalination is still energy-intensive and hazardous to the environment. It consumes 0.4% of the global electricity, that is, 75 TWh per year and also produces 76 million tons of CO_2 annually.[39] Therefore, three major challenges for desalination technologies are (1) high specific energy consumption (SEC), (2) CO_2 emissions from burning of fossil fuels, and (3) negative impacts on marine ecosystems due to the discharge of concentrated brine back into the sea. These challenges propelled the development and commercialization of nanomaterials for respective desalination technologies. Desalination technologies are either pressure-driven, temperature-driven, or chemical-driven processes.

Thermal Desalination. Thermal distillation is a conventional approach mainly used for treating water with a high level of total dissolved solids (TDS; >45,000 mg/L).[40] Thermal desalination processes such as multistage flash distillation and multiple-effect desalination are energy intensive (overall equivalent electrical energy consumption of ∼15−30 kWh_{elec}/m^3 for a power plant running at 30% efficiency), costly (cost of produced water ∼0.52−1.75 US $\$/m^3$), and hazardous to the environment (CO_2 emissions ∼15−30 kg/m^3 for standalone operation and 8−16 kg/m^3 for cogeneration operation).[41] An emerging alternative process is membrane distillation, integrated with carbon nanomaterials.[42,43] As an example, a CNF-ceramic nanoporous composite membrane, which has a 10 μm hydrophobic carbon fiber layer with a minimum pore size of ∼30 nm on a ceramic substrate, has shown greater than 99% salt rejection and 3−20 times higher water flux than traditional polymeric membranes.[44] Hydrophobicity of the CNF layer ensures smooth permeation of water vapor across nanopores, and thermal conductivity enables more than 80% recovery of the latent heat.

Thermal desalination plants running on solar and geothermal energy sources are being explored. Also, the efficiency of solar-powered thermal desalination is being enhanced by improving the performance of solar concentrators by using nanomaterials that have high photothermal conversion efficiencies and energy storage.[45] Nanofluids with extraordi-

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nary thermal conductivity and absorption-emission properties have improved the performance of thermal collectors.[46]

Membrane-Based Desalination. Membrane-based separation is adequate for treating water with TDS typically below 45,000 mg/L.[40] Such technologies (RO, reverse osmosis; forward osmosis, FO; electrodialysis, ED; nanofiltration, NF) have gained immense commercial success due to their much lower specific energy requirement (3−8 kWh_{elec}/m^3), cost effectiveness (0.26−0.54 US $\$/m^3$), and low CO_2 release (1.7−2.8 kg/m^3 for seawater RO) into the atmosphere.[39,41] RO has now surpassed thermal technologies in the desalination market, replacing them as convention. The membrane module is the most energy-intensive part of the desalination process and constitutes nearly 71% (2.5−4 kWh/m^3 for seawater RO) of the SEC.[47] While a majority of RO systems have achieved an SEC of 2.3 kWh/m^3, thermodynamic limit stands at 0.76 kWh/m^3, for a feed having a TDS of 35,000, indicating scope for improvement. However, taking into account an energy recovery of 50%, the practically achievable SEC limit rises to 1.06 kWh/m^3.[48] Reduction in recovery percentage can improve SEC, but will enhance operational and capital costs. The next-generation membranes should therefore focus to overcome the trade-off between permeability and rejection and improve selectivity.

Among membrane materials, nanocomposite polymeric membranes are commercially successful due to their low cost and feasibility for large-scale manufacturing. Polyamide membranes are widely used due to their high selectivity and permeability compared to conventional cellulose acetate-based membranes. Polyamide membranes are composed of an extensively cross-linked nonporous polyamide layer supported by a porous polysulphone layer at the bottom.[49] Progress in the development of membrane materials has been gradual, as permeability and selectivity have to be counterbalanced and fouling probability needs to be decreased. Nanomaterials designed at the molecular level are essential for addressing these challenges. In one such attempt, researchers created a three-dimensional (3D)-printed polyamide membrane prepared by layer-by-layer electrospinning in order to achieve reduced thickness (minimum thickness ∼4 nm) to maximize permeance and increased smoothness (roughness ∼2 nm) to decrease the probability of fouling, while maintaining the membrane's strength.[50]

Membranes composed of aligned CNTs are suitable for ultrafiltration. To improve their selective nature, CNT tips are functionalized with zwitterionic species or aliphatic groups such as carboxylic acids.[51] Integration of CNTs with existing membranes has rendered them superhydrophilic, improved their permeation and solute rejection, increased their lifespan, and led to better electrical and mechanical properties.[52] Another promising class of materials includes graphene and two-dimensional (2D)-derived frameworks that physically separate undissolved solids at the nanometer scale.[53] Lab-scale results indicate up to 1000 times better permeability for graphene compared to conventional thin-film composite polymers as RO membranes.[54] However, scaling remains a challenge. Defect-free aquaporin-based membranes, prepared by embedding bacterial aquaporin Z (AqpZ) into a chemically and mechanically stable matrix of a block copolymer or a unilamellar lipid, match the single-channel water permeability coefficient of conventional polyamide membranes (5−36 molecules s^{-1} Pa^{-1}).[55] Aquaporin-based membranes can potentially achieve a permeability of 601 $L{\cdot}m^{-2}{\cdot}h^{-1}{\cdot}bar^{-1}$, exceeding the performance of commercial RO membranes by 2 orders of magnitude.[56]

Further, Kevlar aramid nanofiber (KANF)-based membranes are an emerging category, constituted of nanoscale form of poly(paraphenylene terephthalamide).[57] Nanomaterials under this class are mechanically robust, flexible, tunable in pore size, electrically conducting, and physically stable. Membranes based on KANFs and their composites have demonstrated over 96% rejection of Rhodamine B dye and Au NPs (∼6 nm).[58] They have also shown a desalination efficiency of 99.7% for Na_2SO_4 by performing electrodialysis-based ion separation at a constant voltage of 15.0 V.[59]

RO can also be made operationally efficient by improving the quality of feedwater with FO pretreatment. Using FO in concert with RO reduces the fouling probability of membranes, decreases the consumption of chemicals used for cleaning, and enhances recovery. However, RO-FO hybrids are practical only above a threshold flux of 30 $L/m^2/h$.[60] Therefore, FO membranes for achieving threshold flux are being developed by incorporating nanomaterials to improve hydrophilicity (TiO_2, halloysite nanotubes, graphene oxide, *etc.*), resulting in a faster transport of water molecules. Loading metal−organic frameworks (MOFs) into existing membranes has shown up to 72% improvement in the permeability of pure water for FO desalination.[61] Another aspect of improvement lies in the fabrication process of commercial polyamide membranes. Structural characteristics of the film, such as, morphology, uniformity in chemical composition, and roughness directly affect the membrane's performance.[62−64] Polyamide thin-film composite membranes, created by conventional interfacial polymerization technique, offer a limited scope for optimization of permselectivity. During polymerization, the ultrafast reaction between *m*-phenylenediamine and trimesoyl chloride monomers causes the system to quickly reach the gel point, thereby limiting any further diffusion of the monomers. This creates selective layers with high heterogeneity in depth and restricted control over film thickness. It can be overcome by performing molecular layer-by-layer deposition (mLbL) which results in relatively smooth films, tunable at molecular level.[65−67] Controllably reducing film thickness helps to minimize pressure requirements, ultimately making RO energy efficient. Amidst these advantages, there still remains the challenge of scalability and limited throughput associated with mLbL techniques.[68]

Chemical Desalination. Of the chemical-driven desalination technologies (*i.e.*, ion-exchange, liquid−liquid extraction, and precipitation), the ion-exchange process is the most commonly used. Ion-exchange uses only 7.2 MJ/m^3 of specific energy while producing CO_2 below 0.7 kg/m^3, compared to RO, which uses 29.5 MJ/m^3 of specific energy and emits 3.8 kg/m^3 of CO_2.[41] Ion-exchange processes are composed of ED and capacitive deionization (CDI).

ED is among the most extensively researched and commercially successful electromembrane desalination method at present. Oxide NPs (SiO_2, TiO_2), carbon nanomaterials (*e.g.*, CNTs, graphene-based nanomaterials), and Ag NPs, zeolites, *etc.*, are examples of nanomaterials that are incorporated into ion-exchange membranes for ED to tune properties such as surface area, ionic conductivity, tensile strength, energy efficiency, thermal stability, *etc.*[69] Fouling, separation efficiency for different types of ions (monovalent, divalent, *etc.*), lifetime, and eco-friendly routes of synthesis are

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current performance gaps of ion-exchange membranes that need to be improved.

Discovered more than 50 years ago,[70] capacitive deionization is promising due to its relatively low input capital, high-energy efficiency, scalability, and minimal maintenance requirements, despite being limited by feedwater's TDS and by capacity. Therefore, variable capacity CDI units with nanomaterials incorporated for effective treatment are expected to be an emerging direction for desalination. Currently, commercial CDI units are available for desalination of brackish water (up to 3000 TDS).[71] Further modification of electrode materials has involved integration with ion-exchange membranes (membrane CDI; MCDI), resulting in up to 80% energy recovery during the regeneration step.[72] Researchers have improved the porosity of CDI electrodes by incorporating oxide NPs (TiO_2, SiO_2, *etc.*) and carbon-based nanomaterials (CNTs, CNFs, graphene, *etc.*), leading to a higher degree of hydrophobicity and enhanced surface area.[73] The MCDI technology utilizing ion-exchange membranes faces challenges of intermittent sequential operation, limited adsorption capacity due to plate electrodes, and expensive CDI cells, which are barriers in scaling up of the technology. Substituting stationary electrodes with flow electrodes consisting of suspended carbon powder in conventional MCDI has enabled researchers to overcome the operational limitation by eliminating the discharging cycle, enabling self-regeneration, and improving the ion adsorption capacity, providing practically unlimited surface area for ion adsorption.[74,75]

ATMOSPHERIC WATER HARVESTING

The earth's troposphere contains approximately 1.42×10^{19} liters of water in the form of water vapor, and the world population today is about 7.6 billion. Therefore, there is nearly 1.8 billion liters of water available per person in the atmosphere. Atmospheric water harvesting, thus, has vast potential, even if only a miniscule fraction of this resource is used. Note that the oceans of the planet were once dry and were filled by rain.[76] Thermodynamics suggests that for an open water surface to attain maximum entropy and equilibrium, water vapor above the surface has to attain saturation, thereby causing replenishment through greater evaporation. We note that excessive extraction of vapor locally might affect the hydrological cycle negatively.

An early example of fog harvesting practice includes one in 1969 in Mpumalanga, South Africa, where two large (~28.0 m × 3.6 m) nets made from plastic mesh were used to harvest fog water for South African Air Force personnel. These nets harvested an average of 11 L/m^2/day during the 15-month interval from October 1969 to December 1970.[77] In another instance, implementation of fog collection took place in a village named Chungungo in North Chile in 1987; 75 nets, each of which were 12 m × 4 m in size, delivered an average of 33 L of CW per capita per day to 330 villagers. Both of these harvesters were passive as they did not require energy input.

Alongside passive water harvesters, systems of active water harvesting that require external energy to produce CW now exist. Active harvesting mechanisms have been translated to commercial atmospheric water generators (AWGs). These AWGs extract moisture primarily by condensation or adsorption mechanisms, or a combination of both. The energy efficiency of AWGs (amount of energy consumed per liter of water generated) renders them fit for regions with relative humidity (RH) > 40%. Hence, AWGs have proven to be a viable alternative in coastal regions where a lack of sufficient resources has deterred the installation of desalination plants. However, they are inefficient when RH drops below 40%.[78] Efficient water-capturing mechanisms are being developed and demonstrated by studying natural phenomena and mimicking them at the micro- and nanoscale.

Identifying natural harvesting routes used by several plant and animal species, understanding their harvesting mechanisms, and mimicking them have helped scientists to create a next generation of nanoengineered materials and structures that harvest moisture and efficiently transport the condensed water. Examples of a few successful mimics include those inspired from the elytra of a Namib Desert's beetle,[79] Stenocara, spider's silk,[80] and banana leaf.[81] Successful mimicking of the surface structures of these species followed by systematic water-harvesting experiments have led to a better understanding of the science of dew condensation, involving the nucleation of droplets and their coalescence and subsequent transportation from the surface.[82−86] Most modern-day condensation-based AWG devices are partially or completely based on the vapor compression refrigeration cycle. Implementing nature-mimicking structures in existing and future AWG devices will boost these devices' ability to operate efficiently, even in harsh, hot, and dry climatic conditions. Micro−nano hierarchical structures have been reported to be the most efficient in terms of offering a large number of nucleation sites as well as fast coalescence and transportation of the nucleated droplets from the surface.[87,88] Sarkar *et al.* recently reported further advancement in this direction with a harvesting efficiency of 56.6 L/m^2/day at 87% RH.[33] We expect that the future of surface science and engineering for atmospheric water harvesting will focus on combining nanoengineered structures with unique wetting gradients. Biomimicked structures can particularly help the AWG market flourish in arid North African and Middle East countries, specific regions of which are severely suffering from water scarcity today. Stand-alone AWG systems working on renewable energy sources should prove extremely valuable to societies residing near coastal regions, where the atmosphere is relatively rich in moisture and electricity costs are too high to afford any other water-delivery solution.

Condensation being infeasible below 40% RH has accelerated the development of materials for adsorption-based harvesting. Conventional desiccants have low absorption capacities (silica gel and zeolites) and slow kinetics (hygroscopic salts) or need energy-intensive regeneration (polymers). An emerging class of MOFs offer high surface area and tunable pore size and hydrophilicity. Their promising performance (2.8 L/day at 20% RH corresponding to a kg of $Zr_6O_4(OH)_4(fumarate)_6$) and unique sorption behavior have shown potential for harvesting in deserts.[89] However, substantial work is required on optimizing thermodynamic properties of MOFs and their variants, along with device engineering, before field implementation. For a detailed discussion on all aspects of atmospheric water harvesting, readers are requested to visit ref 78.

AFFORDABLE NANOSENSORS AND CATALYSTS FOR CLEAN WATER

Although conventional analytical methods such as high-performance liquid chromatography (HPLC) and inductively

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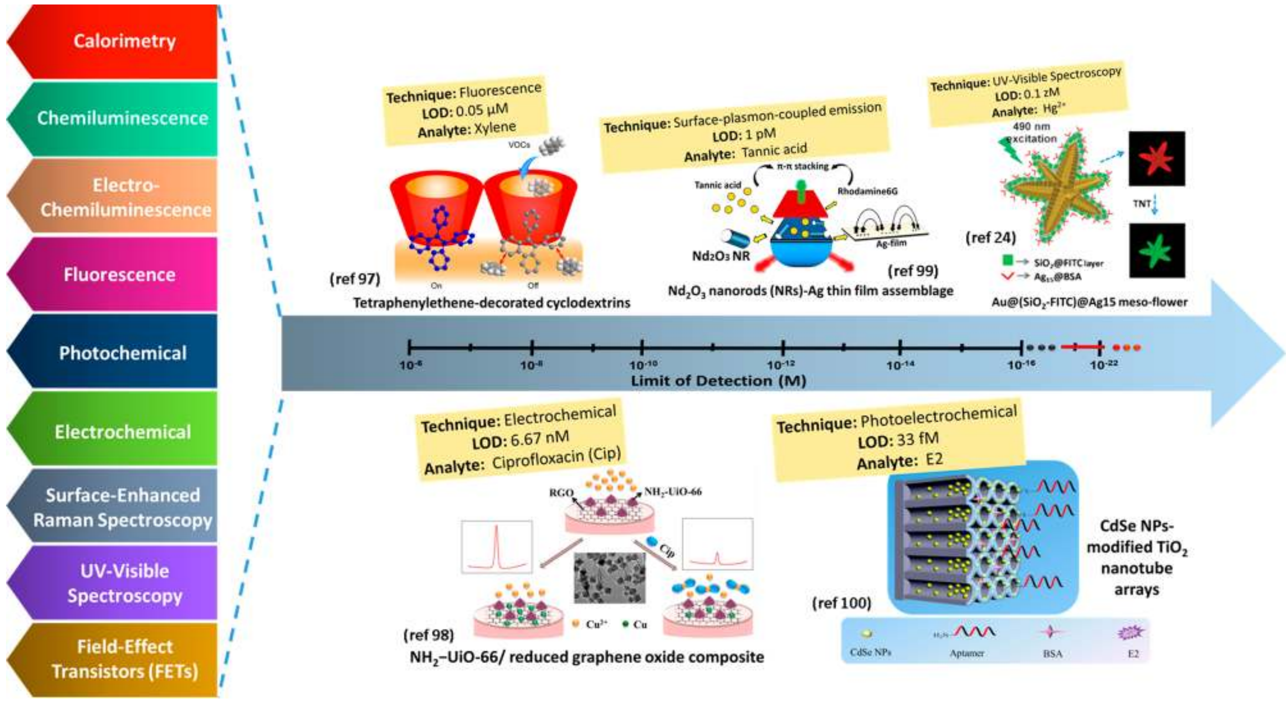

Figure 2. Schematic of the limits of detection (LOD) achieved using nanomaterials. The rightmost part represents detection up to the single-particle/ion level. Reprinted with permissions from ref 24. Copyright 2012 John Wiley & Sons; ref 97. Copyright 2016 American Chemical Society; ref 98. Copyright 2019 American Chemical Society; ref 99. Copyright 2019 American Chemical Society; and ref 100. Copyright 2014 American Chemical Society.

coupled plasma mass spectrometry (ICP-MS) exist for lab-scale testing, high cost, elaborate sample preparation, and unavailability at point-of-use have been limiting factors for their utilization. Nanomaterials are unique in their properties such as optical absorption and emission, which are extremely sensitive to surface functionalization and local environment. These properties have been extensively used in sensing in the context of CW.

An absorbed photon causes several consequences in a semiconducting NP, most important is the creation of a free electron and a hole, both of which can diffuse to the surface of the particle and react with adsorbed water molecules.[90,91] The hole, therefore, can create an oxidizing species such as $HO^{\bullet}$ and the electron can form OH^-, particularly for a hydrated particle in water. Other similar species that can arise are $O_2^{\bullet -}$, $HO_2^{\bullet}$, and $O^{\bullet}$, which also appear on hydroxylated particles. A TiO_2 NP generates reactive species upon photoirradiation, creating an active reaction center that is regenerative in nature, and it becomes the basis for efficient photocatalysis. Consider the following example of photocatalytic sensing of a dye, rhodamine B (RB), using NPs. RB is a contaminant found in wastewater produced from the textile, dyeing, and plastic industries, with proven effects of carcinogenicity, and reproductive and neurotoxicity, thus posing a serious threat to humans and animals upon reaching the groundwater and other water bodies. Photodegradation of an aqueous solution of 10^{-5} M tetraethylated RB in the presence of 100 mg of TiO_2 NPs in a 50 mL solution can be observed visibly upon solar irradiation, with 560 nm light.[92] The self-photosensitized dye reaches an excited state upon visible light absorption, enabling electron injection from the excited state of the dye to the conduction band of TiO_2. Adsorbed O_2 on the TiO_2 surface takes up the injected electron to form $O_2^{\bullet -}$. Protonation of $O_2^{\bullet -}$ forms $HOO^{\bullet}$, followed by further reaction with a trapped electron to form $HO^{\bullet}$, which ultimately leads to degraded products.

Researchers are actively investigating emerging materials and their properties, such as luminescence and catalysis of noble metal clusters, in the context of sensing. Atomically precise clusters of noble metals are composed of a few tens of atoms, with precise composition.[93] One such example is $Ag_{29}(BDT)_{12}(PPh_3)_4^{3-}$ (where BDT and PPh_3 are 1,3-benzenedithiol and triphenylphosphine, respectively), which is intensely luminescent in the red region of the electromagnetic spectrum.[94] Luminescence, as in the case of molecular systems, is extremely sensitive to the medium. Such clusters may also be sensitive to the metal core because they are reactive as well. The core, being accessible to ions and molecules in the medium, makes this chemistry fundamental to developing cluster-based molecular sensors. The catalytic processes and their high reactivity can be used for the destruction of refractory organics.

The luminescence of clusters can be enhanced by anchoring them on plasmonic particles through a process called metal-enhanced luminescence.[95] This enhancement is also possible by embedding clusters onto electrospun fibers.[96] In both cases, it is possible to detect and to quantify contaminants such as mercuric ions down to a few ions, at the single-particle or single-fiber level. Such sensor mats could make test strips affordable for ultrasensitive detection. Figure 2 represents illustrations of detection limits achieved using nanomaterials such as tetraphenylethene-decorated cyclodextrins,[97] NH_2-UiO-66/reduced graphene oxide composite,[98] Nd_2O_3 nanorod-Ag thin-film assemblage,[99] CdSe NP-modified TiO_2 nanotubes arrays,[100] and Au@(SiO_2-FITC)@Ag_{15} meso-flowers,[24] and their corresponding properties.

Achieving higher sensitivity and creating a compact and cost-effective sensor accessory will enable the integration of

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Table 1. Nanotechnology for Clean Water: Status of Implementation[a]

technology umbrella/ref	material	problem addressed	organization	status	cost (cents/gallon)
adsorption/110	FeOOH	arsenic	IIT Madras	commercial	0.14
adsorption/111	nanoalumina fibers	submicron and colloidal particles, bacteria, viruses, dissolved salts, endotoxin, pharmaceuticals	Argonide	commercial	0.03
sorption/112	nanocellulose	metal ions	UPM-Kymmene Oy	commercial	–
adsorption and physical separation/113	zeolite	heavy metals and ammonia	KMI Zeolite	commercial	–
FO/114	CNT-based membranes	brines and several industrial salts	Porifera	commercial	–
RO/115	CNT-based membranes	dissolved salts	NanOasis	commercial	–
FO and RO/116	Aquaporin water channels	micropollutants, xenobiotics, organics	Aquaporin	pilot	–
CDI/36	high surface area carbon electrodes	dissolved salts and metals	InnoDI and IIT Madras	commercial	0.5
NF/117	hollow fiber membrane	salts, heavy metals, toxic chemicals	De Mem Ltd. and NTU Singapore	pilot	–
abiotic chemical reduction or anerobic biodegradation/118	iron nanoparticles	heavy metals, nitrates, phosphates	NANOIRON	commercial	–

[a]Only select technologies and solutions are listed, based on publicly available information. CNT = carbon nanotube; FO = forward osmosis; RO = reverse osmosis.

such devices with smartphones and will facilitate point-of-use applications. A geographical map of contaminants in water could be possible, and it could potentially even be dynamic, like weather maps. An affordable sensor put on all CW sources and service lines will enable continuous monitoring and rectification wherever needed, also verifying the sustainability of the solutions. Upon linking sensors to smartphones, a much-needed in-depth knowledge of global and local water quality will be available as and when needed, which will help to create a rapid action force solving water quality issues. The data generated may provide direction to water quality professionals and help to allocate resources for every region, ensuring the availability of CW for all.

TOXICITY

The properties that make NPs useful and relevant in CW applications can also make them objects of suspicion.[101] Being in the same size regime as biomolecules, NPs can mimic biomolecules and enter biological systems, such as humans, animals, and plant cells, or organelles. Coupled with the possibility of appropriate functionalization, this probability gets further enhanced. When polystyrene NPs (~40 nm in diameter) are adsorbed on medaka fish eggs (*Oryzias latipes*), the NPs reach the yolk and gall bladder during embryogenesis. Exposing an adult medaka to a 10 mg/L NP solution caused NP accumulation in the gills and intestine, with NPs also propagating to the liver, testes, and eventually to the brain through the blood–brain barrier.[102] In light of such findings, NP release in the CW stream has to be controlled, particularly when loose NPs are used in the process. Even in the case of supported NPs, depending on the strength of anchoring, the particles may get dislodged, especially in forced flow. It is also possible for NPs to enter the medium when the surface binding group or ligand is affected by external stimuli such as light. For example, a hydroxyl radical can react by breaking the bond between the particle and the anchoring ligand. One approach to safety is to ensure that the NPs used are similar to natural materials and that their bulk counterparts are available in nature. The only point of concern will be whether the NPs are released in the processed water stream.[103] Evaluating the presence of free particles in water at low concentrations may pose a challenge for measurement. However, this challenge can be solved by understanding the dynamics of release, which depend on factors such as water chemistry, size of NPs, surface area, surface functionalization, *etc.*[104] Particle release may be evaluated using single-particle ICP-MS to track decreasing particle diameters or increasing constituent elements' concentrations as a function of time. One such study highlights the release of silver ions from silver NPs in river and lake water, suggesting more than 80% dissolution for smaller particles (<10 nm), while only 50% dissolution for larger particles (50 nm), over a period of four months.[105]

COMMERCIALIZATION, BUSINESSES, AND INCUBATION

Globally, the water sector is too broad to estimate its net worth. Over the years, nanotechnology-based businesses have emerged in the CW sector to address global and local needs (Table 1). As an example, a lack of freshwater in rural areas has led to the development of rolling water purifying drums, which purify the water collected from a distant place, as one rolls it home.[106] This local solution would prove extremely valuable to people who walk long distances carrying cans of water over their heads and shoulders. Such drums could contain nanomaterials to enable purification during transport. Later, an oil–water emulsion can be prepared by simple agitation, and nanomaterials can be extracted by trapping them at the oil–water interface.[107] Such simple methods of extraction suggest the possibility of using rolling water purification for contaminant-specific treatment in a particular area, by mixing with a specific nanomaterial.[108,109]

Several limitations still exist in the commercialization of NTs for CW, including low governmental investment in and lack of adequate focus on water-related research activities, especially in developing countries. These limitations have resulted in slow progress in the translation of developed technologies. In many ways, this lack of progress is understandable because water availability and associated issues have been major challenges, and therefore investments for

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Table 2. Examples of Emerging Nanomaterials That Could Translate or Have Already Translated into Nanotechnologies for Clean Water

material	application	organization	ref
metal−organic framework	atmospheric water harvesting	Massachusetts Institute of Technology, United States	123
organic-templated nanometal oxyhydroxide impregnated with silver nanoparticles	water purification	Indian Institute of Technology Madras, India	124
cationic and anionic membranes	capacitive deionization	Idropan Dell'orto Depuratori Srl, Italy	125
metal oxide nanocomposite heterostructure powder	sensing of hydrogen sulfide	Honeywell Romania SRL, Romania	126
layer-by-layer assembly of graphene oxide membranes	water purification	University of Maryland, United States	127
aromatic diimide chromophores	sensing of volatile organic compounds	Jawaharlal Nehru Centre for Advanced Scientific Research, India	128
doped carbonaceous material	photocatalytic removal of chemical/biological pollutants and micropollutants	University of Arkansas, United States	129
graphene oxide	dehydration using vapor phase separation or perevaporation	The University of Manchester, United Kingdom	130

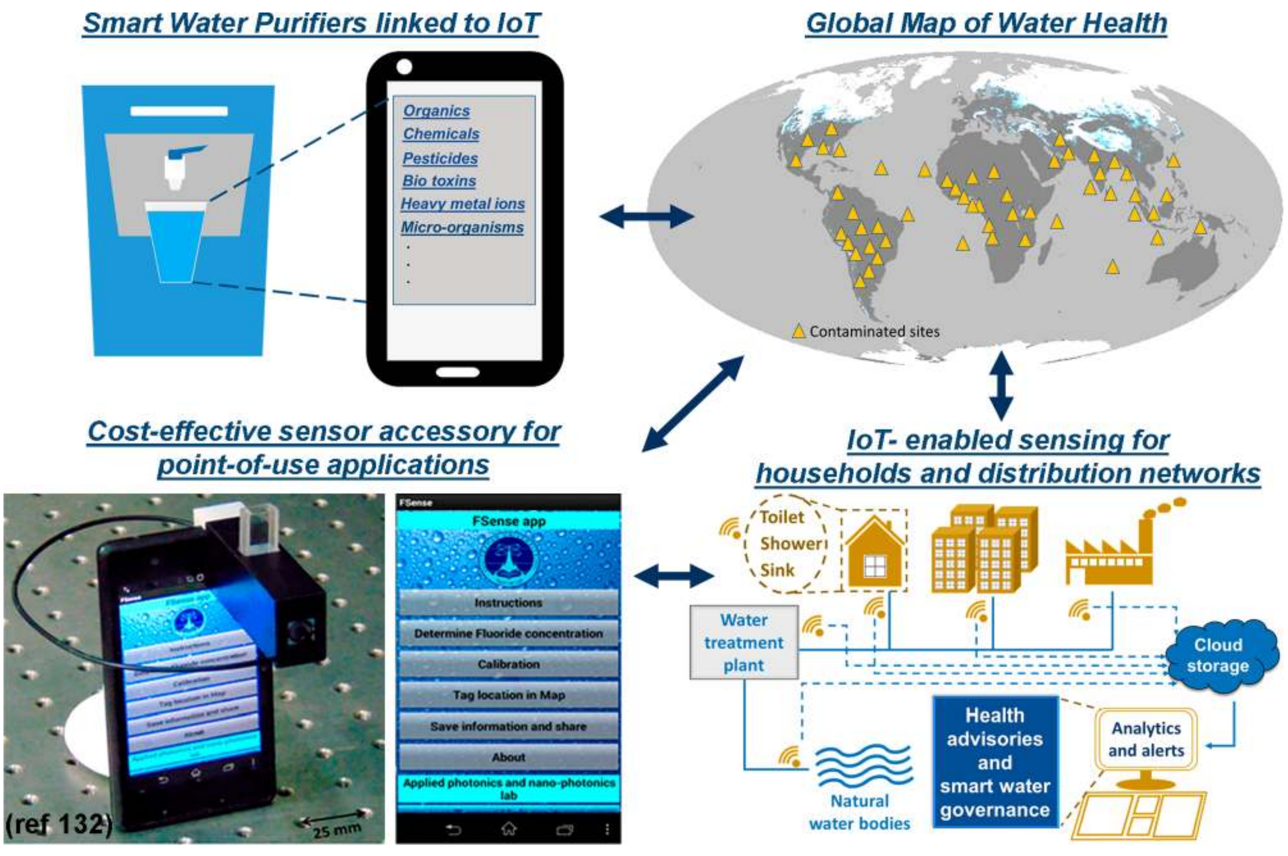

Figure 3. Schematic representation of future data on water being collected from water purifiers, field samples, and city infrastructure through nanosensors embedded in smartphones and IoT-enabled domestic water purifying systems and water distribution networks across the world, proving health advisories in the long run. Top-right panel image adapted and modified with permission from NASA Earth Observatory. Copyright 2020 NASA Earth Observatory. Bottom-left panel image reprinted with permission from ref 132. Copyright 2017 American Chemical Society.

them took priority over research on CW. For example, $0.26 billion were allocated by the Indian government in 2017, under the National Rural Drinking Water Program to provide arsenic- and fluoride-free drinking water to 28,000 habitations in India, which was about 0.007% of the GDP of the country.[119] While allocation to the drinking water segment declined from 87% to 31% in 10 years, the share of rural sanitation increased from 13% to 69%.[120,121] Therefore, 0.1% of the country's GDP, utilized by the ministry in 2018−2019,[122] was majorly expended for combating fundamental challenges of open defecation, improvement in cleanliness, and uniform sanitation coverage, which of course are essential. However, looking at the long-term challenges, a continuous focus of ministry on research on arsenic and fluoride, and related issues would have would have partially solved the problem of these persistent contaminants. Currently, research on CW and implementing solutions for CW are subjects of separate ministries and, therefore, are separately budgeted and administered. Similar situations exist in other countries too.

Table 2 provides illustrations of emerging nanomaterials in the diverse areas of CW that are either serving the community or have the potential to do so.

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WATER PURIFIERS OF TOMORROW

Increasing awareness of the need for essential minerals in water and the dangers of harmful ones will necessitate ensuring that optimal mineral content is delivered through drinking water. Next-generation technologies that can retain certain minerals or reject others completely would make it possible for water purifiers to select purification technologies according to need. All of these in conjunction with Internet of Things (IoT)-enabled devices and the proliferation of Internet availability across the world would enable acquisition and transfer of water quality data across time through personal electronic devices.[131−133] Big data analytics would thus help create personal health advisories. The availability of such data across a population would be of use to communities and governments to understand and to plan for the health of their people. Water purifiers may become intelligent devices in the foreseeable future, as shown in Figure 3.

Future CW solutions will need to be implementable both locally and nationally. The decentralization of CW technologies is essential for any country, but especially for emerging economies. Many nations have adequate resources to empower local governments with region-specific solutions. The decentralization and implementation of technologies will also trigger the generation and employment of local manpower, which would help strengthen the economy, if carried out nationwide. It is vital for forthcoming technologies to be environmentally friendly with no net carbon emissions in order to restore the purity of the planet and sustain its natural resources.

OPPORTUNITIES IN SUSTAINABLE AND AFFORDABLE CLEAN WATER

The global water crisis is being countered today by effective removal of contaminants, creation of robust water networks, real-time monitoring of water quality, and linking these efforts with social, political, and economic action. In this section, we highlight both untapped water resources and major problems in the CW sector where NTs could be useful. We offer a few proposals in the context of Bangalore, India, although any city may be chosen in its place.

(1) Ideally, the world must run with net-zero carbon emissions, converting CO_2 and H_2O to fuels and back again to the same amount of CO_2 and H_2O. This cycle is upset when more CO_2 is produced over time, as we do not know how to fix the imbalance using only sunlight as an energy source. We also do not know how to burn fuels efficiently to produce contaminant-free CW that can be used directly. Perhaps engines of the future can be designed to produce usable liquid water. Note that the unsustainable release of CO_2 into the environment has led to the emergence of CO_2-conversion techniques such as photo/electrochemical reduction, sequestration, *etc.*,[134] powered by renewable energy sources such as solar energy, although they do not perform as efficiently as plants. The burning of octane, represented by the reaction, $2C_8H_{18} + 25O_2 \rightarrow 16CO_2 + 18H_2O$ suggests the formation of 162 g of water per 114 g of fuel or 1.42 g of water per gram of fuel. It might be possible to trap this water, similar to trapping CO_2. India consumed approximately 24 billion kg of petrol in the year 2016−2017,[135] which corresponds to burning nearly 21 billion kg of octane (considering the octane rating as 87), making 186 billion kg of water in a year. Urban water collected this way could grow vegetables on windowsills, as one of the end uses.

(2) Bangalore is meeting nearly 52% of its water requirements by exploitation of its groundwater, through borewells that currently reach depths close to 2000 feet in several parts of the city.[136,137] The rest of the water is supplied by the Arkavathy and Cauvery rivers. Dependence on deep borewells will continue to increase with the city's burgeoning population. Hence, restoring groundwater is a serious challenge. Out of the 33 billion cubic feet of rainwater available annually to Greater Bangalore, 5−10 billion cubic feet can be collected and used to replenish depleted groundwater levels.[138] Lakes and ponds can be rejuvenated using rainwater and can be directed for domestic nonpotable usage. Sewage treatment and use of recycled water will also prove to be vital. In a typical residential building in Bangalore having 500 people, 70% of the total domestic water requirement can be reduced through recycling of greywater, saving ∼ US $14,500 annually and also substantially reducing dependence on groundwater.[139] Decentralization of greywater treatment could be a vital measure to reduce the overall water demand. In comparison to conventional centralized treatment systems, which require a large initial investment on infrastructure and technical manpower for maintenance, affordable decentralized treatment systems can be built using nanomaterials-based strategies. As an example, catalytic oxidation processes involving nanomaterials (ZnO, TiO_2, CNTs, *etc.*) mineralize and partially oxidize organic pollutants into harmless products while also destroying pathogenic micro-organisms to an extent;[140] such processes are scalable toward designing compact decentralized systems. Harvesting atmospheric humidity in highly water-stressed regions of the city will assist as well. Innovative methods of water conservation and recycling which reduce consumption have to be rewarded. Water recycling at the household-level (rapid micro- or "nanorecycling") calls for developing technologies not only in remediation methods but also in enhancing eco-friendliness of materials, in general.

(3) Every personal activity has an impact on water. Water audits on materials of consumption, such as detergents, clothes, food, packaging, paint, furniture, *etc.*, need to happen, and each of them has to be reinvented to make cities livable. It is worth recalling that cotton became water-intensive due to the dyeing industry, which used synthetic dyes during the process of industrialization. Variants of cotton that did not need dyeing, such as yellow and red, were replaced with white by the 1900s, as it could be dyed better.[141,142] Reintroducing these native varieties would reduce dependence on synthetic dyes and detergents.[143] Nanotechnological solutions in the dyeing and leather industries can reduce water consumption and pollution as well.[144]

(4) The maintenance of water infrastructure has caused a rise in the price of CW. According to a 2017 water affordability assessment, the percentage of U.S. households that find water services unaffordable is expected to rise from 11.9% in 2017 to 35.6% in 2022.[145]

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Atmospheric water harvesting and capacitive deionization integrated with next-generation nanomaterials offer affordable solutions, and such technologies, free from municipal water networks, may be a way forward in select areas. Municipal water systems have to be upgraded too, with reduced resistance to flow using NT-enabled coatings.[146]

(5) Bottled mineral water sales continue to rise. A report found the presence of microplastics in mineral water samples in glass and poly(ethylene terephthalate) packaged bottles.[147] However, the effect on human health of such microplastics, additives, and pigment particles of sizes below 5 μm remains unexamined. Integration of nanosensors with smart water bottles and linking of water quality and quantity to an individual's physiological information in real-time has the potential to revolutionize personal health. Patients suffering from kidney diseases and congestive heart failure, with recommended protocols on water intake and its quality, could enormously benefit from such sensors. In addition to such nanosensors, biodegradable materials are needed as a replacement for nondisposable plastics, which could bring about another materials revolution.

Among other synthetic matter is an ever-expanding class of per- and polyfluoroalkyl substances, which comprise nearly 4730 commercially available synthetic chemicals and polymers.[148] A majority of them have high water solubility and mobility and are bioaccumulative in nature. Their sources include manufacturing facilities, industrial and domestic wastewater treatment plants, landfills, *etc*. Given their ability to persist indefinitely in the environment due to the presence of strong carbon–fluorine bonds, they pose a serious threat for future generations, if they reach groundwater through one of the sources. Several nanoenabled remediation strategies have been found promising for these contaminants.[149] Techniques such as electrochemical oxidation and hydrolysis using CNTs, photocatalytic decomposition using nanostructured oxides, reductive degradation using nanoscale zerovalent iron, *etc*., have proven as effective remediation strategies in near-ambient conditions. However, it is imperative that no toxic metal ions should release into the treated water. Immobilization of effective nanosorbents into a matrix could result in a deployable remediation device.

(6) Providing CW for all citizens drains resources initially, but builds resources in the long run. In countries such as India, old practices and emerging aspirations coexist. Villages live on traditional agricultural practices such as burning harvested fields prior to sowing and flooding them during farming. These practices contribute to smog and reduce water availability in neighboring cities, which in turn respond by conveying water from further away, leaving the farmlands dry and inhabitants hungry. Technologies will need to address imbalances of many kinds.

(7) Challenges of CW are linked to clean air, clean energy, sustainable agriculture, and a clean environment. As an example, it is possible to harvest energy from natural and wastewater by utilizing salinity gradients. A power of 0.8 kW/m^3 can be generated by utilizing the osmotic pressure difference between river water (0.01 M NaCl) and seawater (0.1 M NaCl).[150] Globally, an untapped amount of nearly 1000 GW and 18 GW of energy is available, from rivers and wastewaters going into the sea, respectively.[151] Currently, pressure-retarded osmosis and reverse electrodialysis have emerged as membrane-based techniques for osmotic energy harvesting. Existing challenges of having to use expensive materials and the requirement of high power density could be overcome by channelizing energy from complementary sources such as waste heat and brines.

The foregoing suggests that only integrated water management with outside-the-box thinking can make cities breathe better. In the context of overall water balance, we list 10 challenges or opportunities that can be addressed through NT (combined with other technologies) for sustainable cities:

(1) Global CO_2 emissions due to desalination were nearly 76 million tons (MT) in 2015,[39] and global methanol requirements that year were approximately 75 MT.[152] Nanotechnology-assisted production of methanol from CO_2, supported by renewable energy sources, is a promising direction to address both concerns simultaneously. One potential pathway is to develop an efficient catalytic system for CO_2-to-methanol conversion that demonstrates high selectivity, conversion efficiency, and low global-warming impact through the use of renewable energy.[153]

(2) Efficient water-harvesting mechanisms that do not require additional energy input are needed, such as solar-heat-enabled atmospheric water capture by a porous MOF (801, $Zr_6O_4(OH)_4(\text{fumarate})_6$) at a relative humidity as low as 20%.[89]

(3) Nanomaterials can be used to conserve CW by improving the physicochemical and biological characteristics of soil. For example, the application of biodegradable nanohydrogels enhances the moisture content of soil and its water retention capacity, thereby relieving water stress.[154] Groundwater demand by the irrigation sector in India is expected to increase from 605 billion cubic meters (BCM) in 2000 to 675 BCM in 2025, but may be reduced to 637 BCM by 2050.[155] The projected decline in groundwater consumption beyond 2025 is attributed to NTs that can enhance the efficiency of groundwater-driven irrigation.

(4) Water audits in developing consumables from food to toiletries are needed. For example, cradle-to-grave life cycle assessments of the process of washing 5 kg of laundry (requiring medium hardness water at 40 °C and consuming 120 g of liquid detergent, 49 L of water, and 0.53 kWh of electricity per washing cycle) reveals a primary energy footprint of 6.57 MJ equivalent, a carbon footprint of 0.54 kg CO_2 equivalent, and an environmental footprint of 3.34×10^{-2} EI99 points (see ref 156 for a detailed description of the units).[156,157] This understanding may change the consumer's choice of detergents, packaging materials, chemicals, building materials, *etc*. (see point 7 below). The "water positive" aspect of nanomaterials in this context, indicating net CW production during a synthesis, was shown when an antimicrobial silver-based composition was synthesized for controlled release of silver ions. The synthesized material consumed only 1 L of water for its production, while it helped to make 500 L of CW.[158]

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(5) Point-of-use water recycling products for personal and local reuse, such as portable, chemical-free, ozone-based disinfection solutions, using hydrodynamic cavitation, acoustic cavitation, and electrochemical oxidation, may lead to energy-efficient water treatment and recycling across oil and gas industries, municipalities, mining industries, *etc.*, at capacities as large as 12,492 L/min.[159] Note that more than 80% of wastewater is discharged into surface water bodies in developing countries today. India alone generates approximately 6.2 million m^3 of untreated industrial water every day.[160] Such solutions will contribute to better wastewater management and preservation of freshwater resources.

(6) Placing compact nanosensors on water bottles and other water-based beverage containers to monitor water quality (pH, hardness, turbidity, *etc.*) and to create an interconnected network (Internet of Nano Things) will generate opportunities.[133]

(7) Self-cleaning fabrics lead to reductions in consumption of water, detergent, electricity, or an equivalent amount of CO_2. Nanomaterials such as SiO_2 NPs, CNTs, TiO_2 NPs, *etc.*, are known to demonstrate photocatalytic self-cleaning through the creation of hierarchical structures, whereas adsorption of organic molecules such as alkanethiols and fluorosilanes imparts water repellency to surfaces by lowering their surface energy. A U.S. study found that a treated, self-cleaning fabric could reduce electricity and water consumption by as much as 84%, compared to an untreated fabric, while undergoing 50 laundry cycles in its lifetime.[161]

(8) Waterless vacuum toilets with incorporated fecal and urinal waste-repellant nanocoatings are possible at the domestic level.[162] Also, the possibility of nanoenabled nutrient recovery from human feces and urine could be explored for reuse at homes.[163] Overcoming the social stigma is crucial to implement such solutions, particularly in developing countries.

(9) Next-generation membranes for desalination are needed. At present, production of 1 m^3 of CW through RO desalination consumes approximately 3−5 kWh of electricity, although enhanced efficiency has been demonstrated.[164] Highly selective membranes can filter chlorine and boron from seawater in a single pass, unlike the multiple passes required in RO that currently makes desalination a costly, and energy- and time-intensive process. One possible direction is the development of aquaporin membranes that offer the required selectivity. However, efficient salt rejection and cost-effective upscaling of the process are required. Synthetic water channels, mimicking aquaporins, such as CNTs and several aligned peptides to form pores, is another possibility. Tunability in terms of pore size is required here. Other promising materials include graphene oxide,[164] MoS_2,[165] *etc.* The development of next-generation membranes could contribute to achieving the set target of reducing desalination costs from US $2.00 to US $0.50 per m^3 (as per the U.S. Department of Energy).[166]

(10) Nanomaterials can enhance oil recovery in oil and gas industries. For instance, oil viscosity can be reduced by use of a suspension of Al_2O_3 NPs in distilled water or brine, and rock wettability can be modified by the application of silane-treated silicon dioxide NPs. Both of these modifications improve oil recovery.[167] An oleogelator-impregnated cellulose pulp effectively recovers oil from oil−water mixtures by congealing it within a matrix, thereby offering an eco-friendly, cost-effective, and practical solution to restore the marine ecosystem from oil spills.[168]

CONCLUSIONS AND PROSPECTS

CW production presents questions of clean manufacturing, responsible use of materials, equitable distribution, and, ultimately, concern for humanity. Genuine concern for water availability puts limits on reckless growth and consumption. Water, therefore, presents an appropriate subject on which green chemistry and green manufacturing converge for social good. In this Review, we presented directions for materials science and sustainable growth around water, through the eye of NT, which is interlinked with other disciplines. Similar approaches need to be pursued for other areas such as agriculture, energy, housing, and healthcare for a sustainable planet. Nanotechnology can acquire a significant role in all of them as it expands the limits of materials and functions.

For a more comprehensive treatment of the topic with available NTs currently being explored, it is necessary to go through other perspectives and reviews[169−177] as well. Limitations of space and the nature of this review have kept our discussion focused on affordable and sustainable NT. We have taken a significant number of Indian examples in the article with the notion that it is an aspiring country in dire need of generating data on a large number of water-related issues, thereby offering endless opportunities of discussion and work, and the outcomes are applicable across the world.

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Notes

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VOCABULARY

hydrophobicity, a physical property in which molecules show absence of affinity toward water and prefer nonpolar interactions with molecules of similar nature, leading to positive change in their free energy, thereby causing

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segregation of water molecules; **photocatalysis**, a phenomenon in which a reaction is accelerated due to the presence of a catalyst which generates electron−hole pairs upon irradiation to create active species such as free radicals, enabling secondary pathways and reducing activation energy barrier for the reaction; **desalination**, a process of removal of dissolved minerals and salts from high-salinity water to obtain product freshwater and concentrated brine for disposal; **nanosensors**, a class of devices that utilize unique properties of nanomaterials to detect and quantify events occurring at nanoscale; **specific energy consumption**, a variable defined as amount of energy consumed per unit of production

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