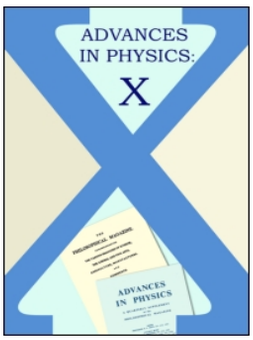

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REVIEW ARTICLE

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DNA-based nanowires and nanodevices

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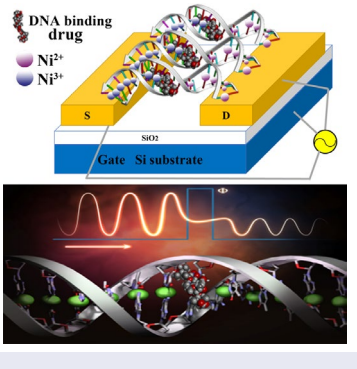
ABSTRACT

DNA (deoxyribonucleic acid) is a highly versatile biopolymer that has been a recent focus in the field of nanomachines and nanoelectronics. DNA exhibits many properties, such as high stability, adjustable conductance, vast information storage, self-organising capability and programmability, making it an ideal material in the applications of nanodevices, nanoelectronics and molecular computing. Even though native DNA has low conductance, it can easily be converted into a potential conductor by doping metal ions into the base pairs. Nickel ions have been employed to tune DNA into conducting polymers. Doping of nickel ions within DNA (Ni-DNA) increases the conductivity of DNA by at least 20 folds compared with that of native DNA. Further studies showed that Ni-DNA nanowires exhibit characteristics of memristors, making them a potential mass information storage system. In summary, DNA molecules have promising applications in a variety of fields, including nanodevices, nanomachines, nanoelectronics, organic solar cells, organic light emitting diodes and biosensors.



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1. Introduction

It is well known that the nucleotide sequence of DNA (deoxyribonucleic acid) molecules is the blueprint of life. DNA is a macromolecule consisting of four types of deoxyribonucleotides, termed deoxyadenylate (A), deoxythymidylate (T), deoxycytidylate (C) and deoxyguanosinylate (G), linked together via phosphodiester bonds. Single-stranded DNA molecules with complementary sequences form a double helix via the Watson–Crick base pairing scheme (A-T and C-G pairing). Aside from their role in biology, DNA molecule exhibits intriguing properties, such as electron conductivity, vast information storage, self-organisation and programmability, making it an ideal material in the construction of nanodevices, nanoelectronics and molecular computing. Metal ion-chelated DNA (M-DNA) molecule has been considered as nanowire with attractive conductivity and proved as a memristor system [1]. The possibilities of merging the gap between nano and macrodevices may be achieved with M-DNA devices.

2. Molecular devices

The concept of nanomachines was first postulated by Feynman [2] and was further illustrated by Drexler in the mid-80s. However, the view of 'cabling of atoms to nano actuator' [3] received much scepticism from the majority of the scientific societies, because atoms are highly reactive, and hard to be manipulated and rearranged. Later, this concept was demonstrated by the fabrication of nanoscaled devices by using different materials such as copper, silver and palladium nanowires [4]. It is not surprising because, compared to atoms, molecules are stable and easy to handle. In addition, many macromolecules already exist in nature and can be self-assembled to form a larger structure. Biomolecules, especially DNA, are ideal materials for molecular devices due to their capability of self-assembly into a structure with a specific function or to perform a single task [5].

Molecular devices, such as molecular wires and/or machines, are components at the molecular level that have large surface-to-volume ratios to present sufficient functional properties of machines at nanoscale [2]. Molecular devices are desirable because they can rapidly accumulate and displace electrons/charges within the nanoscale structures, and are sensitive to changes of physicochemical and biological environments. With these characteristics, molecular wires and/or machines resemble electronic memory units. These molecular devices can be made by cost-effective and low-energy technologies and may possibly provide environmentally friendly solutions for future computing technologies. There are two key approaches to nanotechnology, namely top-down and bottom-up strategies. Solid-state nanodevices can be fabricated via the top-down approach using lithography and/or NEMS (nanoelectromechanical system) technologies. Some disadvantages, however, are associated with top-down approaches, including high cost, requirement of smooth surfaces, high time consumption and inadequate channel length [6]. Alternatively, the bottom-up approach is more suitable and

convenient for molecules with the capability of self-assembly to construct nanoscaled devices and machines in both solid and liquid states [7].

2.1. Top-down approaches

Lithography is a mature and well-developed semiconductor technology and has been applied widely to the fabrication of integrated circuits and micro or even nanoelectronic devices. It is a typical top-down approach to generate numerous micro or nanodevices from a bulk silicon wafer [7]. Taiwan Semiconductor Manufacturing Company (TSMC) can manufacture a 16-nm 3D FinFET transistor using the third-generation high–k/metal gate process, the fifth-generation transistor strain process, and the advanced 193 nm lithography [8]. IBM, on the other hand, can fabricate a 5-nm thin-body SOI (silicon-on-insulator) [9]. Silicon nanowire sensors for real-time chemical detection at a scale of 50 nm were fabricated by electron beam lithography and reactive ion etching on silicon [10]. The direct-write electron beam lithography, and inductively coupled plasma-reactive ion etching (ICP-RIE) techniques were employed by Nuzaihan and colleagues for the generation of triangular-shaped silicon nanowires with dimensions of $20 \times 30 \times 400$ nm (width × height × length) [11]. These silicon nanowires were developed to sense biomolecules based on variations in drain current, electrical resistance and conductance on the silicon nanowires. InGaAsP/InP nanowire arrays of 20–40 nm in diameter could be easily prepared by ICP-RIE on a metal-organic chemical vapour deposition (MOVCD) grown wafer [12].

Presently, state-of-the-art optical lithography could generate nanodevices as small as 14–16 nm [13]. However, the development of next-generation lithography techniques is required to make nanodevices beyond this limit. Nanoimprint lithography, for example, utilises electron beams or X-rays to generate nanoscale patterns to a definition of approximately 5 nm [7]. Several drawbacks may be associated with the top-down approaches in the fabrication of nanodevices. First, the physical and chemical properties of nanomaterials used in nanodevices are less understood, leading to unwanted or unexpected responses. Next, the fabrication techniques for nanodevices are not fully defined and developed. Finally, the top-down approaches in nanotechnology are incompatible with the existing semiconductor production process and, thus, the developed nanodevices cannot be mass-produced [7]. In conclusion, less possible mass production, low capability of scale-up, low sensitivity in detecting errors and high cost are challenges in top-down approaches. All of these problems can be overcome by bottom-up strategies with some restrictions.

2.2. Bottom-up approaches

In contrast to top-down approaches, bottom-up approaches are used to build nanodevices atom by atom and/or molecule by molecule. With the integration of

macromolecular chemistry, biochemistry, and physical concepts, various nanomachines, such as those based on silicon nanowires/chips [14], carbon nanotubes [15], polymers [16] and nucleic acids [17], have been created by bottom-up strategies.

The growth of vertically aligned silicon nanowires (39 nm in diameter) on a substrate via the vapour-liquid-solid growth method is an example of bottom-up strategy [16]. With this method, silicon nanowires can be efficiently incorporated into nanodevices. A silicon-based nanoscale memristor was developed by Jo and colleagues that acted as a synapse in a neuromorphic circuit system to exhibit spike timing-dependent plasticity and offer high connectivity and high density for efficient computing [18]. DNA can self-assemble into specific structures, making it an ideal material for the development of nanodevices. DNA logic gates, for example, were constructed based on the concept of DNA tweezers [19]. The state of the logic gates was determined by the opening and closing of the DNA tweezers, which was controlled by oligonucleotides and restriction endonucleases. With this concept, approximately ten logic gates could be constructed [19]. Although nanodevices can be conveniently fabricated by bottom-up approaches, reproducibility and mass production are the major issues that need to be addressed. Integrating various molecules or building blocks into functional systems is still challenging for bottom-up approaches.

3. DNA-based molecular machines

DNA has already been exploited for the fabrication of nanoscaled systems [20]. Convenient programming of nucleotide sequences and the mechanical rigidity of short double helices of DNA molecules make DNA highly favourable for the construction of nanodevices with designated 1D, 2D or 3D structures. Moreover, DNA molecules can be easily engineered by scientists with atomic precision using DNA manipulating enzymes, such as ligases, nucleases and transferases [21].

DNA origami, a nanoscaled folding of single-stranded DNA via interaction with a large number of short DNA oligonucleotides, has gained much attention recently because of its potential to direct the formation of predefined 2D or 3D DNA structures at the nanoscale [22]. The design and development of 3D DNA origami structures is time consuming and cumbersome. Hence, it is essential to reduce the effort in designing sequences for DNA origami. Many open-source software packages are available to assist the design of DNA origami motifs, such as caDNAno [23], GIDEON [24], NUPACK [25], UNIQUIMER 3D [26] and SARSE-DNA origami [27]. Han and colleagues developed a strategy to design and generate self-assembling DNA origami with defined 3D multifaceted curved surfaces [28]. Round and square-shaped DNA nanostructures of 62 and 85 nm, respectively, were designed. Based on this strategy, a series of DNA origami with high curvature, such as in the shape of hemispheres, ellipsoids, spheres and flasks were also generated [28].

Many applications have been associated with DNA origami nanostructures, including drug delivery, biosensing, nanoelectronics and molecular computing [29–32]. A lithography patterning technology in combination with DNA origami is proposed to facilitate the formation of DNA-templated circuitry structure [30]. A complete nanoscaled circuit is then accomplished by metallising the patterned DNA origami templates to make conductive wires and is integrated with semiconducting materials to provide multiple transistor functionalities. However, correct DNA patterning, DNA metallisation, and the integration between DNA origami and semiconductor materials are challenges that determine the success of this DNA-based nanoscaled electronic circuitry. Interestingly, DNA-based Boolean logic gates can be implemented in a 3D DNA origami box structure [31,32]. The DNA-based logic gate systems are controlled by a set of DNA oligonucleotides that change the structure of the DNA origami and disrupt the fluorescence resonance energy transfer (FRET) between two fluorescent probes attached to the investigated system. The states of DNA-based Boolean logic gates depend on the low or high FRET efficiencies, which are the representative output signals of 0 and 1, respectively [31,32].

Other than DNA origami, several DNA-based nanomachines and nanodevices have also been developed, such as nanoswitches, biped walkers and DNA tweezers. A DNA nanoswitch was constructed by a cytosine-rich DNA strand, which exhibits a pH-sensitive conformational change between a random conformation and an organised conformation called an 'i-motif' [33]. A fluorescent signal could be repeatedly switched on and off upon the oscillation of proton concentrations. A DNA-based nanoswitch with a similar working mechanism was shown to be useful in mapping the spatiotemporal pH changes in living cells [34]. In these cases, the DNA nanomachines were fuelled by protons. DNA nanomachines can also be fuelled by enzymes or DNA [35,36]. For example, one type of DNA tweezers can be operated by DNA molecules [35]. It opens when the 'set' DNA molecule is added, whereas it closes in response to the addition of the 'reset' DNA molecule. An enzyme-operated DNA-switch was proposed recently [36]. Del Grosso and colleagues demonstrated that the opening and closing of a pH-sensitive DNA nanoswitch can be controlled by proton-consuming (glutathione S-transferase) and proton-producing (urease) enzymes, respectively. Interestingly, the applications of DNA nanomachines can be further extended with the integration of proteins or DNA aptamers, a short single-stranded DNA selected for its binding ability to proteins or small molecules. A DNA nanomachine was constructed based on DNA aptamers that specifically bind human blood clotting factor, α-thrombin [37]. This DNA nanomachine can be instructed to bind or release α-thrombin by the addition of operator DNA molecules. DNA-protein conjugates were also widely applied in the development of immunoassays, biosensors, micro-chips and molecular devices [29].

3.1. Conductivity of DNA nanowires

DNA is a perfect nanomaterial for the construction of nanoelectronic devices. However, physically, the DNA molecule is an insulated wire at low bias. Hence, making DNA conductive is essential for the application of DNA in nanoelectronics. Interestingly, low-conductance linear DNA can be transformed into a highly conductive nanowire by doping metal ions or metallisation [38–40]. Metal ions were shown to bind strongly to the base pairs of DNA and are enclosed by the phosphate sugar backbones. In a metal-doped duplex DNA complex, the hydrogen bonds between complementary bases were replaced with metal ions [40]. The duplex DNA can be doped with divalent metal ions at pH ≥ 8.5 by substituting the imino proton of the base in the base pairing [38]. Interestingly, DNA possessed unusual conducting property when it was doped with Co^{2+} or Ni^{2+} ions. By contrast, DNA may be mechanically deformed by doping with silver ions. Based on observations from transmission electron microscopy, scanning electron microscopy and X-ray photoelectron spectroscopy, silver ions were shown to be deposited on the backbone of the DNA duplex, leading to morphological changes and structural rigidity of DNA molecules. With silver ions, the length and height of the DNA were approximately one third shorter and larger than that of those of native DNA [39].

Numerous reports have shown that DNA can be converted into a good conductor by coating with metals, such as silver, copper, gold and palladium. Nanometal wires were fabricated via three methods: (1) Direct assembly, (2) electroless plating and (3) two-step metallisation. In the direct assembly process, positively charged metal ions may bind with the negatively charged back-bone of DNA. The electroless plating method reduces the metal ions to nanoparticles by chelating through nitrogenous bases or phosphate groups of the DNA template. In the third method, reduction reagents, such as aldehyde, were utilised to reduce metal ions, and those metals were then bound to DNA. These methods were utilised to form nanometal wires based on the backbone binding of DNA [41] (Figure 1). Although the deposition of metals on the phosphodiester backbone of the DNA molecule has been well studied, the interconnectivity among metals and the mechanism of conductivity of metallised DNA are still less understood. The conductivity may be formed by transporting charges through the nanometal granules on the surface of the metallised DNA. However, it is not feasible to compare the mechanism of conductivity among metals because of the lack of detailed studies. On the contrary, the binding of Ni ions with DNA base pairs, the corridor of conductivity, and the linkage between metal ions in conducting electrons have been studied comprehensively in recent years [42–44].

3.2. Ni-DNA nanowire characterisation and its conducting mechanism

As mentioned previously, the conductivity of DNA can be enhanced by incapacitating metal ions, especially Ni ions [45]. Among other metals ions, such as

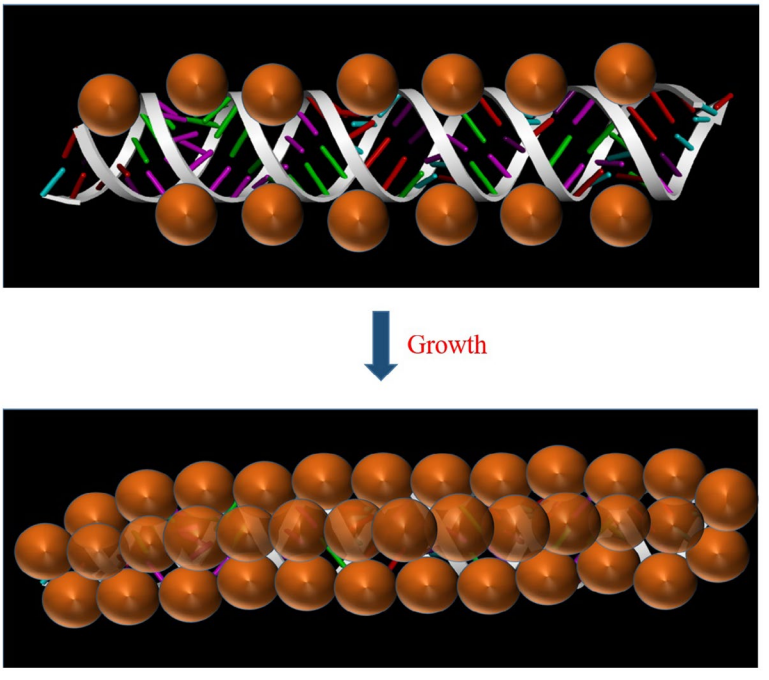

Figure 1. Illustration of DNA guiding nanometal nanowire.
Notes: The DNA is in ribbon and ladder model. The brown spheres denoted the reduced metal nanogranules.

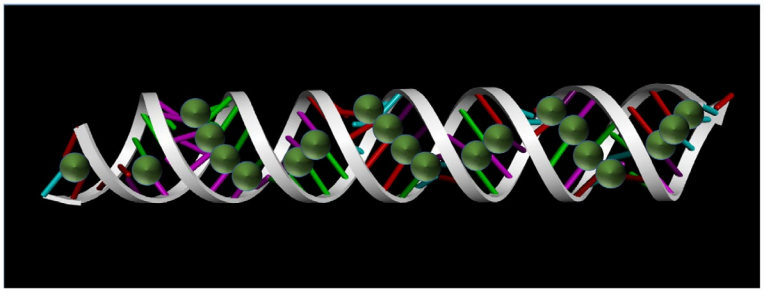

Figure 2. Conformation of Ni-DNA.
Notes: Illustration of the Ni-DNA, where Ni ions are chelated by the base pairs of DNA. Complex of base pairs of DNA (A-T, G-C) chelating with Ni ions act as conducting corridor and phospho-pentose moieties act as insulators in DNA nanowire.

Co and Zn ions, nickel ions provide efficient capacity for DNA to pass electrons through its core. Nickel ion (II) is stable in solution and is able to resist changes in environmental condition. As reported previously, nickel ions can be chelated between paired bases in duplex DNA by replacing imino protons of G and T bases and form stable structures under alkali conditions (pH ≥ 8.5) [38,45] (Figure 2). The bonding of nickel ions with base pairs of DNA was proven experimentally by a reversible chelating reaction, which is based on the fact that the doping of Ni ions may block the access and binding of fluorescent dyes to the major grooves

of DNA [45]. The fluorescence of stained DNA can be recovered by EDTA treatment. Self-assembled monolayers (SAMs) of DNA and Ni-DNA adsorbed on a gold electrode was analysed by electrochemical impedance spectroscopy (EIS) and scanning probe microscopy. The EIS results indicated that the conductance of Ni-DNA was much better than that of native DNA by approximately 20 folds in solution, and Ni-DNA acted as a semiconducting biopolymer with a contact barrier of around 2 eV in Ni-DNA under ambient conditions without excess water [45]. Similar results were also observed by the red-shift in the UV absorption spectra, indicating the bandgap reduction between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of Ni-DNA. Apparently, an electron can be excited from the π state to the π^* state in Ni-DNA with much less energy than that in DNA [45]. These data also imply that the base-pairs of Ni-DNA participate in the charge transport mechanism.

An interesting issue is whether the duplex structure is necessary for the charge transport in Ni-DNA. Cyclic voltammetry (CV) analysis of Ni-chelating single stranded DNA (Ni-ssDNA) and native DNA SAMs indicated that the conductivity of Ni-ssDNA was similar to that of native DNA. Therefore, the duplex form of Ni-DNA is essential for charge transfer. Moreover, the resistance of Ni-DNA polymers increases exponentially with increasing number of base pair mismatches. This data fit well with the Simmons electric tunnel effect model [46,47]. These results indicate that the electrons transport between base pairs with Ni ion chelating follows the tunnelling effect [48]. The relation between the resistance, R, of Ni-DNA and the tunnelling current density J can be expressed as follows:

$$R \propto J^{-1} \tag{1}$$

and

$$J = \left(\frac{e^2}{dh^2}\right)(2m\Phi)^{1/2} V_{\exp}\left[-\left(\frac{4\pi d}{h}\right)(m\Phi)^{1/2}\right] \tag{2}$$

where Φ is the barrier height, m is the mass of the electron, h is Planck's constant and d is the distance between each base-pair. In general, it is around 0.34 Å.

Therefore, the resistance of Ni-DNA is:

$$R \propto \exp(\beta d) \tag{3}$$

where β denotes the constant $(4\pi/h)(2\ m\Phi)^{1/2}$.

Based on the EIS analysis and fitting, the β value is ~0.3 $Å^{-1}$ and the barrier height, Φ, between base pairs is approximately 0.083 eV. Namely, a native DNA with 2000 bp (~680 nm) has a Φ value of approximately 166 eV, with which native DNA may act like an insulator. Apparently, the Ni^{2+}-mediated π-π stacking corridor provides a path for electron transport in Ni-DNA [48].

In order to characterise the charge transport effect of Ni-DNA in a solvent-free system, JangJian and colleagues placed Ni-DNA between two gold electrodes

with a gap of 60–100 nm. In this device, the DNA molecules were stretched by electrophoresis and excess water was removed by baking at 80 °C for 15 min. By applying an external cyclic bias from −10 to +10 V, clear hysteresis in the *I*–*V* curves was observed with two significant negative differential resistance (NDR) peaks at around +3.50 and −4.70 V [44]. This NDR effect denoted that redox reactions of the Ni ions (oxidation of Ni^{2+} and reduction of Ni^{3+}) in the Ni-DNA system are involved. As indicated previously, charge transport through the Ni ion linked the π–π corridor in Ni-DNA. The changes in the redox state of the Ni ions may change the ratio of Ni^{2+} and Ni^{3+} in the Ni-DNA system. It is speculated that the resistance of Ni^{2+} and Ni^{3+} may be different in Ni-DNA [44]. Therefore, the different ratios of Ni ion species in Ni-DNA system may change the effective resistance of the Ni-DNA nanowire device [1]. This external bias-dependent resistance change effect may be recognised as the memory effect of the Ni-DNA system.

This assumption was approved by an assembly with Ni-DNA molecules connected between two gold electrodes using a self-assembly bridging technique developed by Chang and colleagues. This Ni-DNA-gold electrode assembly was visualised and validated by atomic force microscopy [1]. The activity of Ni-DNA as a memristor was demonstrated by its NDR behaviour in response to cyclic-voltage scanning (*I*–*V*). The cycling of switches between *I* and *V* is one of the crucial machineries of memory systems [49]. By shifting the voltage, the number of nickel ion species, Ni^{3+} and Ni^{2+}, in Ni-DNA was altered. The ratio between the Ni ion species in Ni-DNA was not changed without external bias. These observations indicate that the changes in the Ni^{2+}/Ni^{3+} ratio resemble the changes in the writing states of the Ni-DNA memory system, because the redox reaction changes the ratio of the nickel ion species, Ni^{2+} and Ni^{3+}, and the conductance changes consequently. These results imply that the Ni-DNA device can work as a multi-state memory system under various external bias and polarity conditions. Based on these concepts and experimental results Ni-DNA can be exploited as an organic multiple-state information storage device and exhibits high potential to be employed for future nanomachine fabrication.

3.3. Potential applications of Ni-DNA nanowires

As indicated previously, the non-metal chelating sites (base-pair mismatches) in Ni-DNA results in an abrupt increase in resistance. Therefore, the mismatches in the DNA sequences can be detected by doping nickel ions into the short DNA fragments which may be the hot spots of DNA mutations [43]. This working mechanism can also be applied to detect and screen the binding of certain drugs that can interact with DNA.

Interestingly, Ni-DNA can function as a typical nanowire field effect transistor device because it is a conducting nanowire with designable length and conformation. The conducting current through a Ni-DNA nanowire may be altered by the interaction with proteins, DNA, RNA and small compounds [50,51] or by

modifications, such as methylation and damages [52]. Hence, biosensing systems can be developed by integrating Ni-DNA devices for the detection of substances that can interact with DNA, transcription factors or regulators of genes and molecules that cause DNA modification and damages.

Furthermore, the redox states of Ni ions in Ni-DNA can be program controlled by external bias. With this mechanism the writing and reading of information can be done by small external biases and erased by oscillating voltages. Ideally, by controlling the time and polarity of the external bias, a Ni-DNA of 2000 bp in length could have more than 2000 bits of memory states per Ni-DNA unit. A device with 100 units will have 2000^{100} memory states (approximately 1.2676×10^{330} bits). Accordingly, super-high density memory devices can be fabricated by integrating with Ni-DNA nanowires. Furthermore, with matured nucleotide synthesis technologies, micrometre-long Ni-DNA can be easily synthesised and integrated with regular photolithography processes to generate nanoscaled electronic devices.

4. Challenges and issues of DNA nanowires

The development of DNA nanowires has been a recent focus in three aspects: (1) customising the sequence of nucleic acids for better conductivity with reduced mismatching duplex, (2) stacking targeted double-helical backbone for stability and rigidity of nanowires, and (3) interconnection of discrete DNA origami [53]. Even though these targets are achievable, the cost of synthetic DNA and the high error rate of SAMs are problems that need to be addressed in the future. At present, DNA molecules of around 2000–3000 base pairs are utilised to develop model DNA nanowires [1]. To date, the design, synthesis and construction of simple nanowires and nanomachines has been achieved by careful bench-top experiments. However, to move DNA nanowires to bed-side applications, it is necessary to integrate the tiny system with macroscopic structures by organising them on surfaces and/or at interfaces. Finally, DNA can be a nanomachine. To achieve it, the integration of DNA-based circuits with macro machines should encompass rational design, complexity, and stability. The dimensions of the wires will be nanometres in width and micrometres in length. Hence, there is a possibility of cross-conduction of electrons between parallel circuits. This can be avoided by careful design of the circuits and making multiple copies by standard processes. Printing/dipping/nanoimprint and self-assembly of multiple copies of nucleic acids on a chip and/or an electrode will be a typical and cost-effective way to produce standard DNA nanowires.

5. Conclusion and perspective

Nature always creates big things from a dot. Today, we can do anything with a dot having arranged atoms in a distinct manner called a ‘nano-system’, as postulated by Feynman [2]. It is an organic system where each and every part of the terrain

is ruled by organics. The kingdom of 'Artificial Intelligence' was presided by silicon technology and in the future, it may be overtaken by DNA. Nucleic acids have gained importance in biological fields over five to six decades and are now the focus in future electronics. Because of its capability of self-assembly, 1D–3D designable structure, and conductivity inducibility, DNA can be viewed as a nanowire and is proved to be a memory capacitor. The next step of DNA nanowires will be the development of circuiting microchips by connecting and patterning nucleic acids to their terminals. Miniaturised circuits with DNA codings may be the next generation of memory systems. Although multilingual effort from chemistry, computer science, biology and physics is required, the premiere of a DNA nanodevice will be in the not-too-distant future.

Disclosure statement

No potential conflict of interest was reported by the authors.

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