Chapter

Metallic Nanowire Percolating Network: From Main Properties to Applications

Daniel Bellet, Dorina T. Papanastasiou, Joao Resende, Viet Huong Nguyen, Carmen Jiménez, Ngoc Duy Nguyen and David Muñoz-Rojas

Abstract

There has been lately a growing interest into flexible, efficient and low-cost transparent electrodes which can be integrated for many applications. This includes several applications related to energy technologies (photovoltaics, lighting, supercapacitor, electrochromism, etc.) or displays (touch screens, transparent heaters, etc.) as well as Internet of Things (IoT) linked with renewable energy and autonomous devices. This associated industrial demand for low-cost and flexible industrial devices is rapidly increasing, creating a need for a new generation of transparent electrodes (TEs). Indium tin oxide has so far dominated the field of TE, but indium's scarcity and brittleness have prompted a search into alternatives. Metallic nanowire (MNW) networks appear to be one of the most promising emerging TEs. Randomly deposited MNW networks, for instance, can present sheet resistance values below 10 Ω/sq., optical transparency of 90% and high mechanical stability under bending tests. AgNW or CuNW networks are destined to address a large variety of emerging applications. The main properties of MNW networks, their stability and their integration in energy devices are discussed in this contribution.

Keywords: transparent electrode, silver nanowire, copper nanowire, transparent conductive material, stability, percolating network

1. Introduction

Transparent electrodes (TEs) are key components for many industrial devices. TEs indeed do concern applications related to energy field such as photovoltaics or efficient lighting (light emitting diode, LED, or organic-LED, OLED), smart windows or supercapacitors and are therefore associated to rapidly increasing industrial needs. For photovoltaics, the need of TEs concerns, for instance, the front electrode that should be transparent for the sunlight while collecting the photogenerated carriers. For efficient lighting, this is the opposite physical phenomenon: injecting carriers (electrons and holes) by applying a voltage through transparent electrodes to let the generated light exit the LED or OLED device. But TEs are also used in many other applications such as transparent heaters, touch screens, sensors or radio-frequency (RF) devices.

The main TEs investigated in the last decades have been transparent conductive oxides (TCO) [1–4] with the most well-known and used one in the industrial area being indium tin oxide (ITO). And aluminium-doped zinc oxide (AZO) [2] and fluorine-doped tin oxide (FTO) [5] have been also the subject of many studies. While TCO can exhibit good or even very good physical properties, the recent industrial needs have prompted a search of new materials to replace TCO for several applications [6]. Indeed indium, for instance, can be scarce, its deposition often requires vacuum, and TCO by nature are brittle and therefore not compatible with flexible applications. Materials such as carbon nanotubes [7], graphene [8], conducting polymers [9, 10], metallic grids [11] and metallic nanowire networks [12, 13] have been mainly studied for this purpose, and some of them exhibit already promising properties for several applications. In particular, several studies have lately demonstrated that metallic nanowire (MNW) percolating networks can exhibit high electrical conductivity, high optical transparency and high flexibility [12, 14, 15]. The main investigated are silver nanowire (AgNW) and copper nanowire (CuNW). The very high aspect ratio of the nanowires (i.e. length divided by the diameter) allows these networks to achieve very good performances, similar to ITO, however by using much less raw material [12]. Such quantity are often expressed in terms of the so-called areal mass density (*amd*), defined as the required mass of metal (for MNW networks) or indium (for ITO thin layers) per square metre. Their ranges are between 40 and 200 $mg.m^{-2}$ for AgNW or CuNW networks and roughly 750–1050 $mg.m^{-2}$ for ITO thin layers [12]. With rather similar price per unit mass for both In and Ag, replacing ITO by AgNW networks appears to be a cost-effective alternative. Moreover MNW-based TEs exhibit two additional assets: they can be fabricated via solution-based methods, and they present outstanding flexibility (and even good stretchability). These two assets constitute clearly key points for an efficient industrial integration. Another advantage of MNW networks is their high optical transparency in the near-infrared spectrum, especially when compared with TCO: this is of importance for transparent solar cell applications. For those reasons, printed AgNW network-based electrodes have shown a potential as transparent and flexible electrodes in many displays such as solar cells [16–19], OLEDs [20], displays [21], supercapacitors [22], transparent heaters [23–25], radio-frequency antennas [26], antibacterial films [27] or smart windows [28].

In this contribution, we focus on TEs made of AgNWs or CuNWs and will first briefly discuss the role of the nanowire dimensions (both length and diameter) and network density on the physical properties. The network stability will be discussed followed by methods to enhance it, which appears to be a crucial issue for an efficient integration of this technology. Finally, we will briefly discuss the integration of MNW network-based transparent electrodes for energy applications.

2. Main properties of metallic nanowire networks

The main physical properties of metallic nanowire networks are optical properties (transparency and haziness), electrical resistance and mechanical properties (or more precisely the electromechanical properties mainly in bending or stretching modes). These properties depend on several parameters, including MNW dimensions (diameter and length), junction resistance and network density. We will briefly describe this dependence below. **Figure 1** shows single MNW and MNW network observed by electron microscopy at different scales.

MNW dimensions can influence the properties of MNW networks. MNW diameter, D_{NW}, can be first compared with the mean free path of electrons, Λ, in

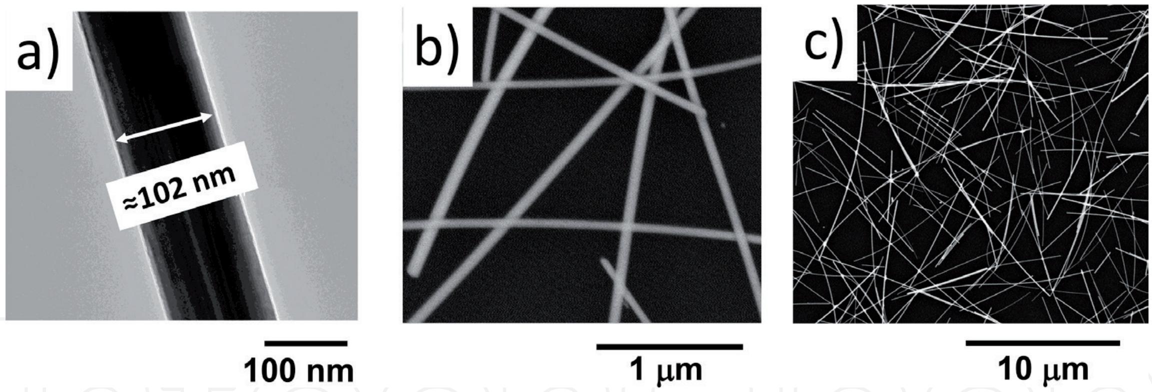

Figure 1.
Electronic microscopy observation of silver nanowires and the associated network. (a) Transmission electron microscopy image of AgNW; (b, c) scanning electron microscopy images of random AgNW network fabricated by spray on glass substrates observed at two different magnifications.

the bulk metal: if D_{NW} is comparable or even smaller than Λ, then surface scattering is increased (for bulk Ag Λ is close to 50 nm). This was derived and observed experimentally by Bid et al. at the individual MNW level [29] and shown to be in good agreement with experiments on AgNW networks by Lagrange et al. [30]. Too small MNW diameter leads to large electrical resistance and to instability at lower temperature [30], while too large MNW diameter increases shadowing effects and then reduces the optical transparency; therefore a trade-off should be found. Also one should keep in mind that large D_{NW} values lead to larger haziness. The influence of MNW length was, for instance, studied by Bergin et al. [31] or by Marus et al. [32]: generally speaking increasing MNW length results in an improvement of their optoelectronic performance. It is also worth noticing that the MNW length distribution can also play a role, as shown by Langley et al. [33], who showed that the critical density of MNW associated to the percolation threshold decreases when the MNW distribution is increased.

The junctions between MNW play also a key role. Recently Ponzoni showed that the relative contribution to electrical conductivity between nanowires and junctions could be very close [34], in good agreement with Bellew et al. who reported junction resistance measurements of individual silver nanowire junctions [35]. Bellew et al. were able to demonstrate, based on experimental data and modelling, that the junction contribution to the network's overall resistance could be reduced even beyond that of the nanowires themselves. It was shown experimentally by several methods that junctions' resistance could be reduced: for instance, Langley et al. showed that a thermal annealing can drastically reduce network resistance thanks to a local sintering of the junctions [36]; and Garnett et al. used light-induced plasmonic nano-welding to *optimize* junction resistance of MNW networks thanks to an efficient *localized* heating compatible with low-thermal-budget substrates such as polymeric substrates [37].

The network density is a key parameter and influences both the optical transmittance and the electrical resistance. Instead of considering the network density (expressed as the number of MNW per unit area), one often prefers to consider the areal mass density, amd, expressed in mass per unit area (mg/m^2). Optical transmittance is observed to decrease linearly with amd as shown by Bergin et al. [31] or by Lagrange et al. [30]. This can be simply explained by shadowing effects [30]. Conversely electrical resistance drastically decreases when amd is increased; therefore an inherent trade-off between high transparency (observed for low amd values) and low resistance (large amd values) has to be considered. **Figure 2a** illustrates the influence of amd value on the electrical resistance. Experiments performed on AgNW

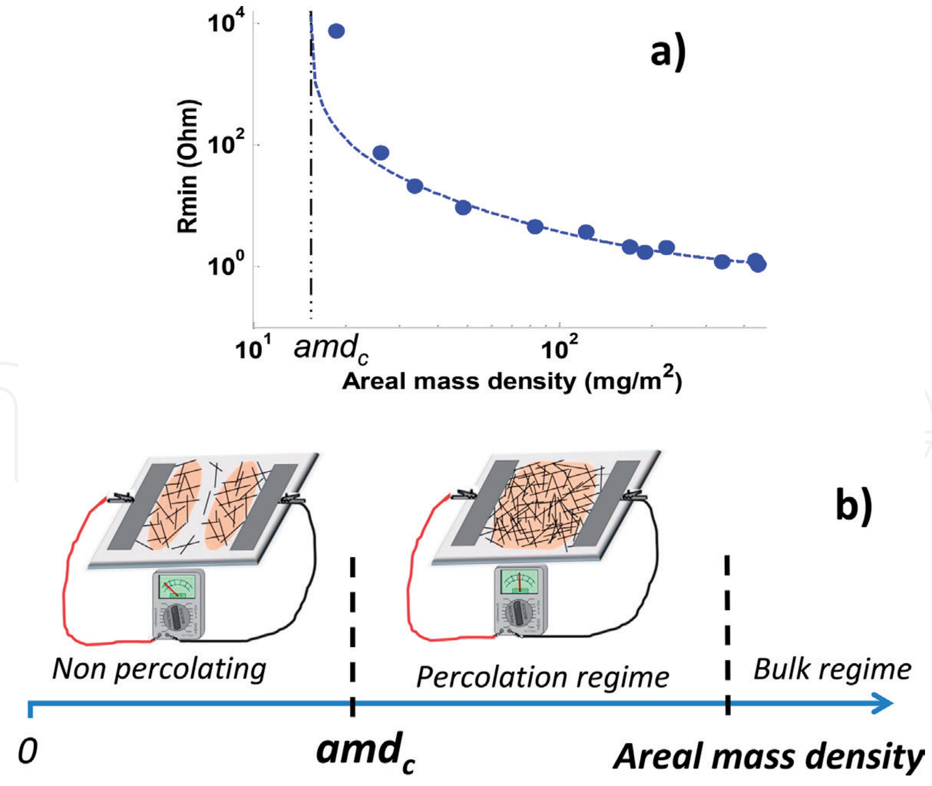

Figure 2.
Effect of the network density, expressed here as the areal mass density (amd), on the electrical properties of percolating AgNW networks: (a) Minimum electrical resistance measured experimentally during thermal ramp versus the network areal mass density [36]; (b) illustration of the three regimes associated with different values of amd. Below a critical value, amdc, no finite electrical resistance can be measured. Just above very large resistance values are measured, while they decrease as a power law for larger amd values; the onset corresponds to the stick percolation. Another transition, less known and studied, exists between the percolative regime and the bulk regime; for the latter the electrical resistivity does not depend anymore on the network density [24].

networks associated to different networks amd values show the existence of a critical value of amd, amdc, below which no finite resistance can be measured (see **Figure 2b**). This limit is associated to the stick percolation, and Monte Carlo simulations show that the amdc value is given by amdc = 5.64 < MMNW>/L2 where <MMNW > is the average mass of the MNW and L is the MNW length [33, 38]. Above amdc the measured electrical resistance is decreasing rapidly following a power law, as shown by **Figure 2a**: there is a rather good agreement observed between experimental data (symbols) and percolation theory (line) [30]. Some differences between real-world networks and Monte Carlo simulations were investigated lately by Langley et al. [33]. The real-world imperfections of a network concern the MNW length distribution, the non-isotropic MNW orientation and the MNW curvature: the influence of these three parameters on the onset network percolation was studied by Langley et al. [33]. For much larger amd value, another transition does exist between the percolative regime and the so-called bulk regime [24]; while this transition is much less known or investigated than stick percolation, such a transition occurs close to amd values that are considered in most applications.

Another way of looking at the influence of amd value on electrical properties of MNW networks is proposed in **Figure 3** where the electrical conductance is plotted versus relative amd. Below the critical amd value, amdc, the experimental resistance is infinite. Just above Sannicolo et al. demonstrated that a discontinuous activation of efficient percolating pathways takes place [39]: experimentally, for sparse networks abrupt drops of electrical resistance are observed. Such an original phenomenon was called 'geometrically quantized percolation' and was observed by lock-in thermography which evidenced the existence of individual hotter pathways

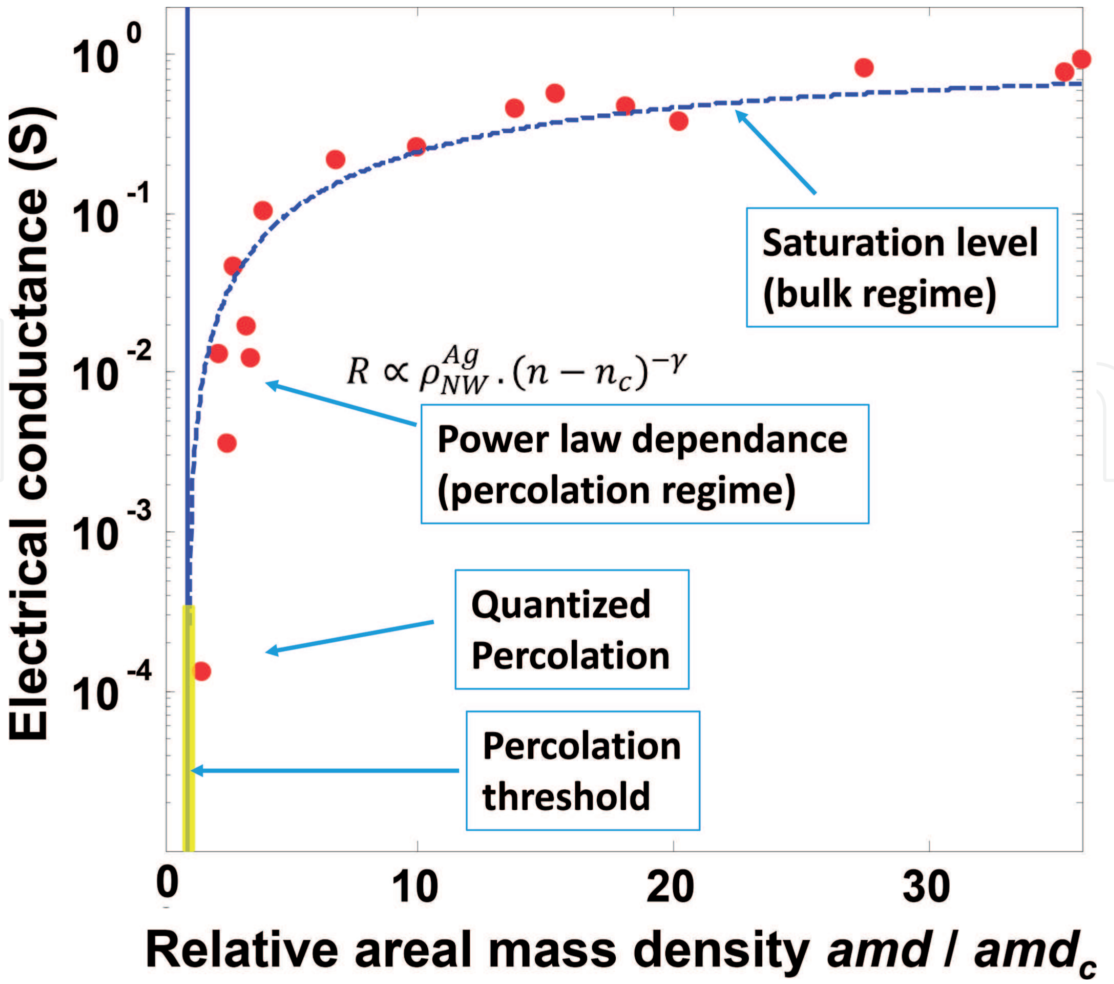

Figure 3.
Electrical conductance of AgNW network versus the relative areal mass density of the network; this shows the different electrical regimes (see ***Figure 2*** *and the text for more details). The red symbols correspond to experimental data, while the blue dash line corresponds to the percolation theory.*

through the network [39]. For larger amd values, one observes the percolative regime for which the electrical resistance is proportional to (amd-amdc)-γ where γ = 4/3, as shown by Lagrange et al. [30]. The previous expression has been used to fit the data of **Figure 3**, and a good agreement is observed for a large range of amd values, while percolation theory should be only valid for amd values slightly above amdc. For very large values of amd, the electrical resistivity does not depend upon amd value, and a metallic bulk behaviour should be then observed.

3. Stability of silver nanowire networks

The stability of metallic nanowire networks appears as a crucial issue, specifically when such TEs undergo thermal and electrical stress [29, 30]. This concerns nearly all applications, and the stability is related to electrical and thermal stability but also long-term ageing and chemical degradation. Such instability can stem from different physical mechanisms such as diffusion of metallic atoms, electromigration processes during electrical stress or oxidation of silver or copper if networks are in contact with either humid atmosphere and/or in high temperature conditions and/or under electrical stress.

One of the first investigations of the instability of AgNW networks was reported by Khaligh and Goldthorpe [40]: they showed that when AgNW-based TEs undergo similar electrical currents than those encountered in organic solar cells, the TEs

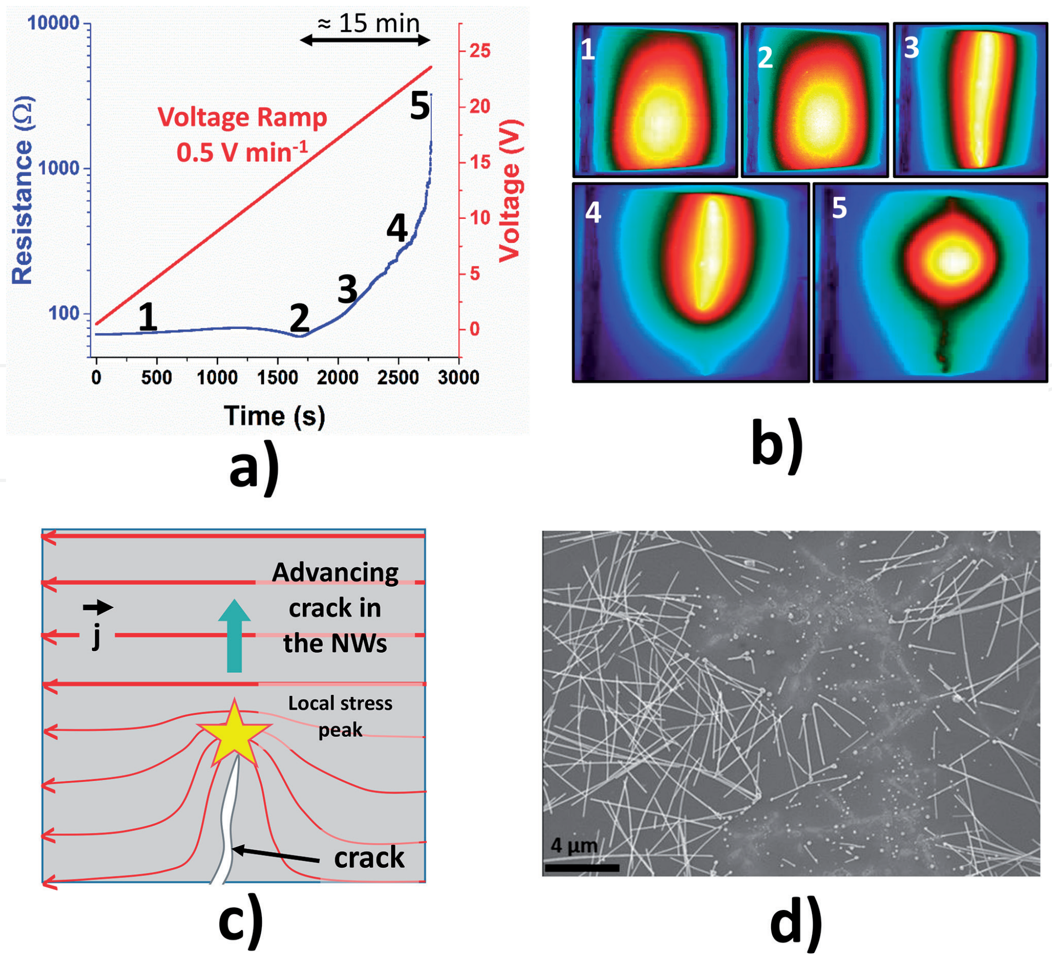

Figure 4.
Electrical failure observations. (a) Time dependence of the electrical resistance of a AgNW network during an electrical ramp of 0.5 V min^{-1}*; the electrical breakdown is observed for voltage larger than 9 volts. (b) Corresponding thermal maps captured in situ with an IR camera at specific times during the experiment depicted in (a) (the electrodes at opposite sides of the specimen are vertical); (c) schematic representation of the mechanisms involved in the crack explaining the crack propagation; (d) scanning electron microscopy observation of the AgNW network location where the crack took place during electrical breakdown.*

can quickly fail within 2 days. They reported that such failure is associated with local Joule heating which deteriorates AgNW network and eventually leads to the network failure. Similar observations were also reported by Chen et al. [41]. Such stability issues are even more pronounced for CuNWs since oxidation (even at low temperature) of CuNWs can occur [42–44].

Figure 4 exhibits the electrical failure observations. **Figure 4a** shows the typical time dependence of the AgNW network electrical resistance during an electrical ramp with the electrical breakdown observed for voltage larger than 9 volts. At first a slight increase of the resistance is observed: thanks to the Joule effect, the network temperature is slightly increased, and since its behaviour corresponds to a metal, its electrical resistance thus increases. For voltage larger than 9 volts, the network electrical resistance drastically increases: this is associated to the electrical breakdown of the network [45].

Figure 4b exhibits in situ thermal maps of the same specimen considered in **Figure 4a** with identical corresponding numbers during the voltage ramp. During the degradation phase (i.e. between (2) and (4)), the heat distribution appears to narrow to a vertical central part of the network parallel to the contact electrodes. At step 4, the accelerated increase in resistance can be associated to the occurrence of a 'thermal' crack which is clearly detectable at the bottom at step 5. A schematic representation of the involved mechanism is shown **Figure 4c**: the propagation mechanism of the crack is related to the displacement of the local current stress peak, which keeps being constricted to the top extremity of the deteriorated area, leading to a runaway-like destruction phenomenon. Finally as shown by **Figure 4d**, AgNWs located close to the crack do appear fully or partially spheroidized, as a result of a drastic local heating and/or electromigration.

To mitigate such instability problems, several studies showed that coating of MNW networks with either inorganic nanoparticles or thin films can drastically improve MNW network stability and integration into real devices. One can cite Morgenstern et al. [46] who showed that solution-processed AgNW films coated with ZnO nanoparticles enhance performance of such TEs when integrated in organic solar cells. The observed efficient protection against AgNW oxidation or degradation leads to a photocurrent for the organic solar cell which appears to be enhanced when compared with ITO use [46]. And, Göbelt et al. showed that a thin aluminium-doped zinc oxide layer deposited by atomic layer deposition (ALD) on AgNW leads to similar photovoltaic performances [47], however by using a much lower silver amount. Some other studies also recently reported that the coating with very thin film layers of titanium dioxide (TiO_2) [48] and zinc oxide (ZnO) [49] clearly enhances the stability. It is worth noticing that new depositing approaches have been assessed lately, and one of the most promising appears to be the atmospheric pressure spatial atomic layer deposition (AP-SALD) [50, 51]. While maintaining the advantages of ALD (viz., low-temperature deposition, thickness control, high-quality materials, and conformity), it can be much faster (up to 2 orders of magnitude faster) than ALD. Another clear asset is its compatibility with roll-to-roll and open air technology. Lately, our team used AP-SALD [52] to fabricate nanocomposite-based TEs in which AgNWs are protected by a conformal thin oxide layer (ZnO). The AP-SALD method was used to deposit thin layers of 15–30 nm ZnO around the AgNWs with the goal of enhancing the network stability [52]. The ZnO coating improved the adhesion of the AgNW networks to the glass substrate, which is known to be poor for bare AgNW networks. **Figure 5** illustrates the positive effects of a coating on MNW-based TE by showing the evolution of the electrical resistance of bare or ZNO-coated AgNW networks during a voltage ramp of 0.1 V/min. A clear enhancement of electrical stability is observed, reducing the degradation of such TEs. As shown by **Figure 5** and by Khan et al. [52], the thicker

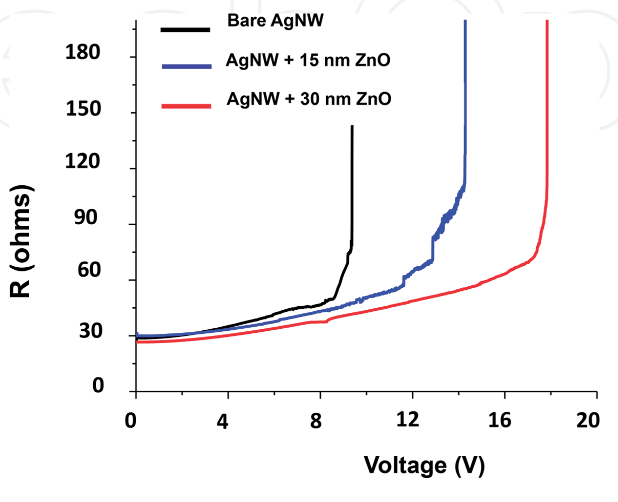

Figure 5.
Electrical failure observations and stability enhancement thanks to coating of MNWs. Variation of electrical resistance for bare and ZnO-coated AgNW networks when subjected to voltage ramps of 0.1 V/min. The bare AgNW network shows failure at around 9 V, whereas the stability of ZnO-coated AgNW networks increases with the increasing ZnO coating thickness to 14 and 18 volts for, respectively, 15 and 30 nm of ZnO coating.

the deposited ZnO layer, the better the stability. This stability enhancement can be explained as follows: the ZnO oxide coating can (at least partially) hinder silver atomic diffusion through the oxide coating [52], avoiding the spheroidization and/or electromigration of AgNWs. A compromise in terms of oxide coating thickness has to be considered depending on the target application since the thicker the ZnO coating, the lower the optical transparency.

Another example of stability enhancement was reported by Shi et al. [53] who demonstrated that transfer of CVD grown graphene onto CuNW films drastically enhances the stability of the hybrid films over long time scale (up to 180 days), while different ageing conditions were also investigated. Graphene is shown to play a key role for preventing oxygen species permeation which drastically decreases oxidation rate. This allows to obtain stable CuNW networks associated with both high optical-electrical performance and excellent stability [53].

In summary to avoid any degradation or oxidation, metallic nanowires are nowadays often coated by a protective layer for an improved integration. This protective (nanoparticles or thin inorganic) layer could be either metallic [54], based on graphene, polymeric or a transparent oxide [52]. This leads in general to a much enhanced thermal and electrical stability along with a better adhesion, although this is at the expense of optical transmittance decrease. One can also observe that conformal thin oxide coating deposited either by ALD or by spatial ALD appears to be an efficient protecting coating while keeping rather high network transmittance [52].

4. Use of metallic nanowire networks for energy applications

Among others, photovoltaic systems, light-emitting diodes (LED) or smart windows constitute sustainable green energy technologies which have been intensively studied lately for energy saving and/or for an alternative to fossil fuel energy. Generally speaking the main goals associated with these technologies concern cost reduction, efficiency improvement and use of abundant materials. For photovoltaic and efficient lighting (LED or OLED), the light should either enter a solar cell or exit the LED requiring the use of an efficient transparent electrode for, respectively, collecting or injecting the carriers. Several investigations have shown that MNW network-based transparent electrodes can be efficiently integrated in such energy devices thanks to their electrical and optical properties. Their excellent flexibility constitutes a clear asset for flexible devices and/or when fast (and then low-cost) technologies such as roll-to-roll are used for the industrial fabrication. And the possibility to coat MNWs allows to tune the work function and band alignments and can therefore lead to better integration possibilities.

For solar cells, MNW networks have been mainly tested in organic solar cells. One of the first demonstrations was reported by Leem et al. [55]: these authors used AgNW network as electrode in P3HT/PCBM organic solar cells, and it showed an efficiency of 2.5% which was equivalent to ITO-based devices. And Yang et al. showed that by using fully solution-processed polymer, bulk heterojunction (BHJ) solar cells with anodes composed of AgNWs were successfully fabricated with performances slightly lower than when ITO is used [56]. Interestingly they showed that the BHJ solar cells were highly flexible since the fabricated solar cells exhibited recoverable efficiency even under large bending deformation up to 120°.

AgNW and CuNW can also be efficiently integrated in OLED devices. AgNW networks were the first to be integrated in OLED devices: The obtained electrode was shown to be suitable for the fabrication of high-performance polymer-based LED [57]. A very recent study by Lian et al. reported the use of CuNW-based composite film for OLED integration [58]. A good electrical conductivity (22 Ω/sq), high transmittance (88%), low surface roughness and good adherence to the substrate were observed. The good adherence originates from the presence of the polymethyl methacrylate (PMMA) coating on CuNWs. The fabricated CuNW/PMMA composite film appears to be stable since it can resist air, water and ethanol exposure without electrical deterioration. With this CuNW/PMMA composite film as anode for the OLED, the device performances appear even better than those with using ITO anode [58].

Several articles clearly also showed that MNW network-based transparent electrodes can be efficiently integrated within electrochromic devices. Thanks to the chrono-amperometry curves and the corresponding in situ transmittance curve at 1100 nm for a WO_3/AgNW film, it was clearly demonstrated that good performances can be obtained when the fabricated electrochromic device uses AgNW network [59]. Such fabricated films exhibit excellent cycling stability as well as distinct modulation of near-infrared light compared with ITO-based electrochromic devices.

Another energy application which appears promising for MNW integration in energy area concerns supercapacitor. Yuksel et al. [22] fabricated nanocomposite electrochromic supercapacitor electrodes: AgNW network electrodes and a green to transmissive electrochromic polymer (PDOPEQ). These authors showed that the obtained supercapacitors have a changing colour from vibrant green to transparent with very good characteristics (i.e. specific capacitance of 61.5 F/g at a current density of 0.1 A/g, capacity retention upon 20,000 galvanostatic charge-discharge cycles). Such characteristics appear very promising for use of MNW-based transparent electrodes in electrochromic supercapacitors [22].

5. Conclusions

As discussed MNW networks exhibit strong potential to act as efficient transparent electrodes for many applications. Indeed, MNW exhibits high transparency and low electrical resistance levels, which are associated with excellent bendability and good stretchability. This contribution reports briefly the influence of main parameters on the MNW network-based TE, the prevailing parameters being MNW chemical nature and dimensions as well as network density. Still, for approaching an efficient integration into industrial devices such as organic solar cells, efficient light production or smart windows, several other requirements have to be considered. One of the most important ones concerns their stability which appears to be a crucial issue: it can involve either electrical, thermal and mechanical aspects or ageing and chemical degradation. The origin of failure in MNW networks was discussed in this contribution with the following stages: optimization, degradation and breakdown of the MNW network. The breakdown occurs via a localized mechanism thanks to the creation and propagation of a crack. To prevent such instability, encapsulation of MNW network is performed by thin oxide layers: this leads to a drastic enhancement of the MNW networks stability. Moreover such approach shows improved adhesion and much better thermal and electrical stability. Finally this contribution shows that the scientific community has worked in several directions and has demonstrated that MNW network-based transparent electrodes can be integrated in industrial devices such as organic photovoltaic, light-emitting diode, in smart windows or supercapacitors. The prospects concern the replacement of AgNW by cheaper MNW such as CuNW while the stability might be a stronger issue than for AgNW, a better optimization of the many parameters (MNW chemistry and dimensions, coating, etc.) for a given application and to make MNW deposition and optimization fast and low-cost enough to be compatible with industrial challenges (for instance, compatible with the very fast roll-to-roll technology).

Acknowledgements

The authors warmly thank Y. Bréchet, C. Celle, D.P. Langley, T. Sannicolo, S. Aghazadehchors and J.-P. Simonato for fruitful discussions. This work was performed within the framework of the Centre of Excellence of Multifunctional Architectured Materials 'CEMAM' n° AN-10-LABX-44-01. This work was funded by the Agence Nationale de Recherche (ANR, France) via the programme ANR-16-CE05-0021 (Despatch), ANR-18-CE09-0041 (Meaning), ANR-18-CE09-0036 (Panassé) and ANR-15-CE05-0019 (Indeed). This work was as well supported by Région Auvergne Rhône-Alpes through the project Pack Ambition Recherche 2018 Eternité. The Carnot Energies du Futur is acknowledged through the project FREE.

Conflict of interest

The authors declare no conflict of interest.

Author details

Daniel Bellet[1*†], Dorina T. Papanastasiou[1], Joao Resende[1], Viet Huong Nguyen[2,3], Carmen Jiménez[1], Ngoc Duy Nguyen[4] and David Muñoz-Rojas[1*†]

1 Université Grenoble Alpes, CNRS, Grenoble INP, LMGP, Grenoble, France

2 Faculty of Electrical and Electronic Engineering, Phenikaa University, Hanoi, Vietnam

3 Phenikaa Research and Technology Institute (PRATI), A&A Green Phoenix Group JSC, Hanoi, Vietnam

4 Département de Physique, Université de Liège, CESAM/Q-MAT, SPIN, Liège, Belgium

*Address all correspondence to: daniel.bellet@grenoble-inp.fr and david.munoz-rojas@grenoble-inp.fr

† These authors contributed equally to this work.

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