

Hybrid Nanomaterials for Flexible Electronics

Materials Engineering / Department of
Mechanical and Materials Engineering

Bachelor's thesis

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15.5.2026

Turku, Finland

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Bachelor's thesis

Subject: Materials engineering

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Title: Hybrid nanomaterials for flexible electronics

Supervisors: Rituporn Gogoi PhD, Aman Kumar MSc(tech)

Pages: 31

Date: 15.5.2026

Research on flexible and wearable electronic devices has been expanding at a rapid pace due to scientific breakthroughs in the development of nanomaterials such as metal nanowires, carbon nanotubes, graphene and MXenes. These nanomaterials exhibit extremely desirable electrical, mechanical and optical properties for the fabrication of flexible electronics. However, each nanomaterial has their own distinct drawbacks and weaknesses, which greatly limit their applicability in certain devices. To combat some of these weaknesses, research on processes where two or more nanomaterials are combined into a hybrid nanomaterial has increased considerably. This thesis introduces some of the more prominent one- and two-dimensional materials with some of the hybrid nanomaterials that have been synthesized. Research on hybrid nanomaterial-based flexible electronics and whether hybrid nanomaterials are capable of replacing conventional conductive materials are considered.

Keywords: one-dimensional nanomaterials, two-dimensional nanomaterials, hybrid nanomaterials, flexible electronics, wearable electronics

Kandidaatintutkielma

Tutkinto-ohjelma, oppiaine: Materiaalitekniikka

Tekijä: Viljami Soini

Otsikko: Hybrid nanomaterials for flexible electronics

Ohjaajat: Tohtori Rituporn Gogoi, DI Aman Kumar

Sivumäärä: 31 sivua

Päivämäärä: 15.5.2026

Taipuisien, joustavien ja puettavien elektronisten laitteiden tutkimus on kasvanut nopeasti nanomateriaalien, kuten metallinanolankojen, hiilinanoputkien, grafeenin ja MXeenien, kehityksessä saavutettujen tieteellisten läpimurtojen ansiosta. Näillä nanomateriaaleilla on erittäin tavoiteltavia sähköisiä, mekaanisia ja optisia ominaisuuksia joustavan elektroniikan valmistukseen. Jokaisella nanomateriaalilla on kuitenkin myös omat selkeät heikkoutensa ja rajoitteensa, jotka voivat merkittävästi vähentää niiden soveltuvuutta tietyissä laitteissa ja käyttökohteissa. Näiden heikkouksien vähentämiseksi tutkimus, jossa kahta tai useampaa nanomateriaalia yhdistetään hybridinanomateriaaleiksi, on lisääntynyt huomattavasti. Tässä kandidaatintyössä esitellään keskeisimpiä yksi- ja kaksiulotteisia nanomateriaaleja sekä niistä syntetisoituja hybridinanomateriaaleja. Työssä tarkastellaan hybridinanomateriaaleihin perustuvaa taipuisaa elektroniikkaa sekä arvioidaan, ovatko hybridinanomateriaalit kykeneviä korvaamaan perinteisiä johtavia materiaaleja tulevaisuuden elektroniikkasovelluksissa.

Avainsanat: yksiulotteiset nanomateriaalit, kaksiulotteiset nanomateriaalit, hybridinanomateriaalit, taipuisa elektroniikka, puettava elektroniikka

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Abbreviations

ITO	Indium tin oxide
0D	Zero-dimensional
1D	One-dimensional
2D	Two-dimensional
CNT	Carbon nanotube
TMD	Transition metal dichalcogenide
TE	Transparent electrode
NW	Nanowire
MNW	Metal nanowire
AgNW	Silver nanowire
AuNW	Gold nanowire
CuNW	Copper nanowire
FCS	Flexible conducting surface
SWCNT	Single-walled carbon nanotube
MWCNT	Multi-walled carbon nanotube
TFT	Thin-film transistor
CVD	Chemical vapor deposition
(r)GO	(reduced) Graphene oxide
h-BN	Hexagonal boron nitride
BP	Black phosphorus
MOF	Metal-organic framework
LED	Light-emitting diode
OLED	Organic light-emitting diode
FET	Field-effect transistor
EDLC	Electrical double-layered capacitor

1 Introduction

The field of electronics has become one of the largest technological frontiers of the modern society, becoming a part of nearly every aspect of human life from transportation and automation to communication and healthcare. During the last two decades, rapid advancements in computing power, efficiency, miniaturization and materials engineering have enabled electronic systems to become increasingly more integrated into everyday environments. As the lives of humans become increasingly more automated through technologies such as the Internet of Things, wearable devices, artificial intelligence and smart healthcare systems, the demand for electronics that are flexible, stretchable, lightweight and non-invasive has grown massively [1], [2], [3].

One of the most prominent developments in the field of electronics is the emergence of flexible devices. Flexible electronics are designed to maintain their electrical properties even while mechanically deformed by bending, stretching or twisting. Multifunctional flexible and stretchable electronics have shown great promise in human health monitoring, biosensors, implantable medical devices, wearable electronics, energy harvesting devices, electronic skins (e-skins), flexible displays, soft robotics and smart textiles [1]. Commercial devices already exist, including foldable smartphones, flexible organic light-emitting diode (OLED) displays, touch screens and wearable fitness trackers [1], [3]. Before any newly developed materials, conventional electrode materials with great conductive capabilities like gold, silver, copper, aluminium and indium tin oxide (ITO) were used in a wide range of electronics. Modern smartphones heavily rely on gold and silver, where they act as electronic circuits and connectors with their excellent electrical conductivity, while ITO is widely used in transparent electrodes and conductive coatings for touch screens [1].

Despite their advantages, the materials mentioned before have shown to have certain problems when applied into flexible electronics. Metals such as gold and silver have excellent electrical conductivity and chemical stability, but are becoming increasingly scarcer and more expensive, limiting their scalability in manufacturing. Copper is notably cheaper than gold and silver while keeping the excellent conductive properties, but suffers from oxidation and stability issues [4]. In addition to these metals, the industry standard ITO has an inherently brittle structure making it unsuitable for most flexible devices [1]. There is also an issue with the need of a large variety of different materials for the different components needed to achieve multifunctionality in applications such as wearable devices. The variety of materials also increases the risk of the

material interfaces being damaged due to differences in mechanical behaviours, leading to poor stability of the device [5].

Due to the limitations of conventional materials, a search for alternatives capable of meeting the requirements of next-generation flexible electronics has begun. Notably, nanomaterials have emerged as great candidates due to their exceptional electrical, mechanical and optical properties that differentiate them from their conventional counterparts. Nanomaterials often exhibit high aspect ratios, meaning a high ratio between the material's longest and shortest dimension, great electron transport properties and excellent mechanical flexibility. These properties enable the creation of a variety of new applications using nanomaterials as the base [1], [3].

Nanomaterials are generally divided to three groups according to their dimensions. These groups include zero-dimensional (0D), one-dimensional (1D) and two-dimensional (2D) structures. Especially 1D and 2D nanomaterials such as carbon nanotubes (CNTs), silver nanowires (AgNWs), graphene, transition metal dichalcogenides (TMDs) and MXenes have shown to possess excellent properties for flexible electronics. Due to their high aspect ratios, these nanomaterials can form interconnected conductive networks using relatively small amounts of material, making it possible to create lightweight and highly conductive nanofilms. These percolation networks also allow these materials to maintain their conductivity even under repeated mechanical deformation and strain [1]. A notable and important property of the multifunctional materials researched for flexible electronics is their optical transparency. Transparent electrodes (TEs) are of great importance in the manufacturing of flexible displays, OLEDs, touch screens and wearable electronics [6].

To achieve even greater efficiency, research on hybrid nanomaterials has emerged as an important subject, because many nanomaterials by themselves rarely possess all the required properties for multifunctional devices. Combining multiple nanomaterials into hybrid structures, it may be possible to eliminate certain weaknesses of one material with the strengths of another, enhancing conductivity, flexibility and mechanical durability. These hybrid structures might make the development of thinner, lighter, more energy efficient and mechanically flexible electronic devices possible [1]. As electronics become increasingly more integrated into wearable and biologically compatible systems, hybrid nanomaterials are expected to be critical in creating the next generation of flexible electronic technologies.

The purpose of this thesis was to conduct a literature review on research published between 2020 and 2026, focusing on one-dimensional and two-dimensional nanomaterials, their strengths and weaknesses, and consider some of the possible hybrid nanomaterials that have been synthesized by combining these nanomaterials. The aim was to evaluate whether the use of hybrid nanomaterials is a viable approach to eliminate some of the weaknesses singular nanomaterials possess in order to create superior multifunctional hybrid systems for flexible electronic device fabrication.

2 One-dimensional Nanomaterials

A requirement of manufacturing flexible electronics is the development of electrodes that can flex and stretch without losing their conductive properties. A strong contender in the flexible electrode field is one-dimensional (1D) nanomaterials. 1D nanomaterials are defined as structures that are in nanoscale in two of the three dimensions, and include transition metal nanowires, metal nanotrugs and -fibers, 1D semiconducting nanowires and carbon nanotubes. 1D nanomaterials are mostly studied as flexible conducting materials due to their excellent conductive properties. These properties stem mainly from their 1D structure, which has a decreased amount of defects, thus creating a minimally resistive path for the transportation of charges [1].

2.1 Metal nanowires

Metal nanowires (MNWs) are considered as metal-based 1-dimensional materials, that have a diameter less than 100 nanometres and an aspect ratio greater than 100. MNWs were originally developed to replace the brittle natured ceramic ITO as flexible electrode materials to make flexible device application more achievable. On top of ITOs' fragile nature, there is also a supply scarcity issue, which limits its use in electronics development and production [6]. The most prominent metals used in MNWs are silver, copper and gold, which all have excellent electrical conductivity made possible by the high density of free electrons [7]. MNWs are used widely in many types of electronic devices, like flexible displays, sensors and field-effect transistors [1].

Perhaps the most promising of the researched nanowire materials has been silver. In addition to silver nanowire's (AgNWs) excellent electrical conductivity and flexibility, a notable strength is their optical transparency. This transparency is achieved due to the nanoscale dimensions of the wires and is an excellent property in optoelectronic applications [1]. Despite their advantages, AgNW applications have been partly limited by their short length (1-20 μm) [7]. In a single line of AgNW, the electrons are able to pass through the single-crystalline structure with low resistance. The main issue with AgNWs are the joint areas where two pieces of wire connect. In the joint areas, the two crystalline structures do not line up, therefore creating a highly resistive junction. Longer AgNWs have been researched to reduce the amount of highly resistive junction locations [1], [7]. Research has also been conducted, where the nanowire

junctions were welded together by thermal annealing, mechanical pressing and plasma treatment [1].

Another metal that exhibits superb electrical conductivity is gold. Gold nanowires (AuNWs) have been utilized in wearable electronics research in the form of Ag@Au core-shell nanowires (Ag@AuNWs), where silver nanowire cores are coated with a layer of gold. These Ag@AuNWs have been researched in stretchable interconnectors and temperature sensors [5]. However, gold and silver are both extremely expensive as materials and thus limits their use in large-scale manufacturing [4]. A cost-effective alternative metal to gold and silver with similar properties is copper. Copper nanowires (CuNWs) exhibit electrical conductivity rivalling that of AgNWs, and as a material, copper is around 91% cheaper compared to silver [3], [4]. Studies have been conducted where CuNWs have been used as flexible conducting surfaces (FCSs), which demonstrated stable resistance even under extreme bending. Like the others, CuNWs also have innate challenges in their application in flexible devices, namely that CuNWs are extremely susceptible to corrosion and other long-term deterioration mechanisms under certain conditions [4].

2.2 Metal nanotroughs and -fibers

Due to limitations of increasing MNWs aspect ratios, research on metal nanotroughs and -fibers has also been conducted. Like MNWs metal nanotroughs and -fibers exhibit great mechanical and electrical properties for flexible electronics [1], [8]. On top of these, metal nanofibers also have a large selection of possible materials, and exhibit possibilities for surface functionalization and porosity tuning. These possibilities enable metal nanotroughs and -fibers to be adjusted according to the application, which makes them ideally versatile. Due to their excellent aspect ratios, mechanical flexibility and sensitivity, metal nanofibers have been used in multiple flexible sensor applications, such as flexible pressure and strain sensors [8].

A fabrication method for the nanotroughs happens with electrospinning process, where an ultra-long polymer nanofiber-based sacrificial web is formed [1]. After that, the metal is deployed on the web, whereafter the polymer nanofibers are selectively removed by dissolving as shown in Figure 1. A similar electrospinning method can be used for metal nanofiber synthesis.

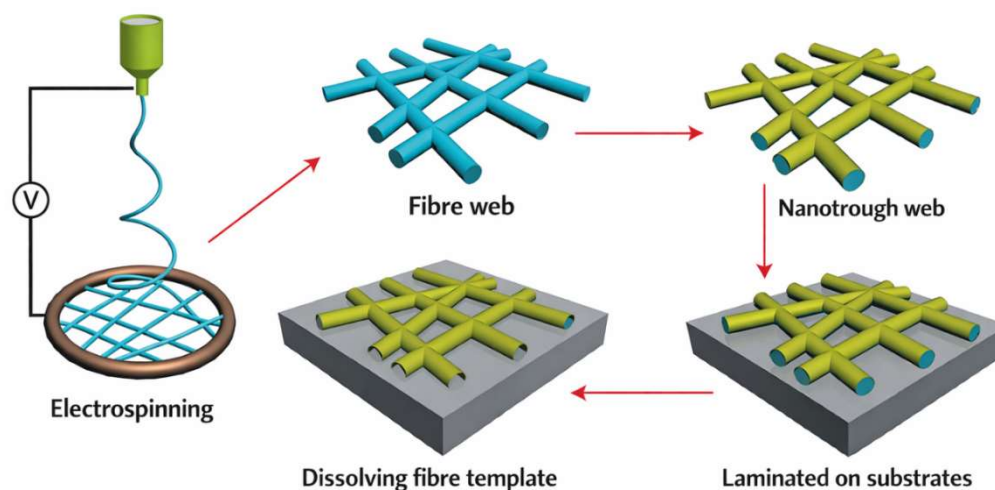


Figure 1. Schematic representation of the polymer-nanofiber templating method for nanotrough creation. Image reproduced with permission from [1], licensed under CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>).

Among other methods, this manufacturing method still requires multiple processing steps that limit its use in large-scale fabrication [1], [8]. The inefficiency and poor yield of 1D nanomaterial manufacturing methods combined with their poor chemical and thermal stability makes the requirement for more in-depth research obvious [8].

2.3 1D semiconducting nanowires

One-dimensional semiconducting nanowires (NWs), unlike their metallic counterparts, are usually made of inorganic materials such as Si and Ge, or from transition metal oxides like ZnO, though organic semiconducting NWs do also exist [9]. The inorganic nature of the materials of these semiconducting NWs allows for more cost-efficient material selection and manufacturing methods, which is an advantage compared to MNWs. Similarly to the metal nanowires, semiconducting nanowires have excellent thermal and electric conductivity, which are of great importance in nanostructures [1], [9]. Inorganic semiconducting NWs have exhibited large aspect ratios and the ability to be dynamically engineered as heterostructures, such as core-shell NWs with other nanomaterials [9].

Conventional NW fabrication processes are usually based on subtractive manufacturing methods, that leads to significant amounts of waste during the process [9]. Additive manufacturing methods for semiconducting NW synthesis has been researched, which would allow for less wasteful and more ecofriendly methods. More eco-friendly methods would lower the raw materials used in manufacturing, while also reducing the cost, making synthesis more lucrative and scalable [9]. The versatility and properties of 1D semiconducting NWs and the

development of more eco-friendly manufacturing methods has made semiconducting NWs a strong candidate for green energy technologies like solar cells, batteries and energy systems for wearable electronics [9], [10].

2.4 Carbon nanotubes

Carbon nanotubes (CNTs) are a larger group of 1D materials consisting of carbon allotrope-based tube structures. Allotropes that can be used in CNTs are graphite, graphene and fullerene. These structures are formed into sheets, that are then rolled up to form tubular structures as depicted in Figure 2. CNTs have an aspect ratio of over 333 and have excellent electrical conductivity, mechanical robustness and chemical stability [1]. CNTs have seen a significant rise in demand now that artificial intelligence and new “smart” technologies have grown in popularity [11].

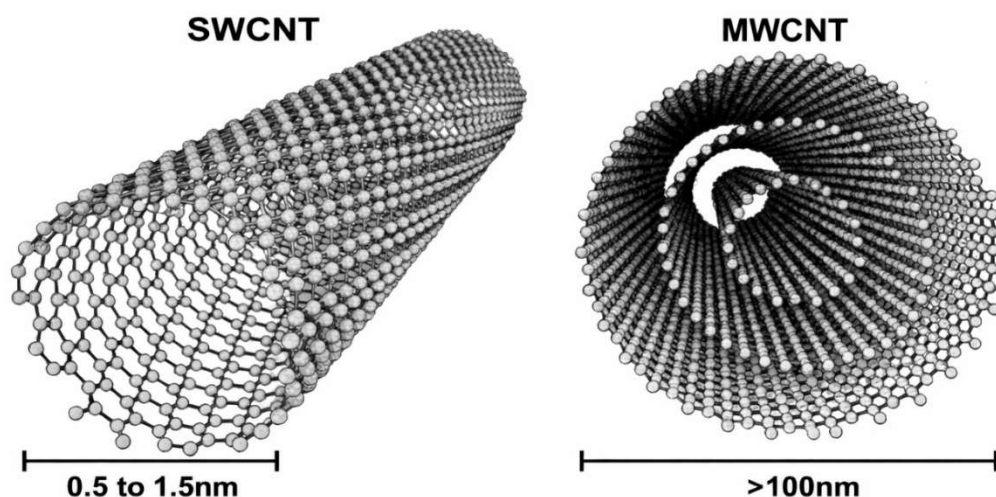


Figure 2. A schematic representation of SWCNT and MWCNT structures. Image reproduced with permission from [12], licensed under CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>).

Structurally, CNTs are usually categorised in two categories, single-walled CNTs (SWCNTs) and multi-walled CNTs (MWCNTs) based on the number of overlaying tube structures [1], [12]. Despite the similarities between the two, SWCNTs and MWCNTs have their own distinct properties, that mostly originate from the multi-layered tube structure of MWCNTs. Because SWCNTs have a single walled structure, they are structurally more flexible than MWCNTs, and have a theoretical Young’s modulus as high as 1 TPa [12]. Due to SWCNTs single-walled structure, they are more complicated to fabricate than MWCNTs and therefore more expensive.

Compared to SWCNTs, MWCNTs have the advantage of a higher aspect ratio due to the multiple layers in the tube. This structure also makes them structurally stronger, but also stiffer and therefore less flexible making SWCNTs more suitable in most flexible electronics applications. Multi-walled CNTs have been especially promising in the fabrication of flexible strain-sensors [13].

Another lucrative application for CNTs are thin-film transistors (TFTs). Conventional TFTs have been mainly based on different types of silicon and organic semiconductors. TFTs based on these materials have kept up with the increasing demand for speed and energy efficiency by their ability to be scaled up in size. However, these TFT could only be improved so far, and eventually hit a limit in their scalability [14]. This limit could only be crossed by changing the materials the TFTs are based on, and thus research on CNT based TFTs begun. CNTs are a suitable replacement due to the need for sub- 10 nm technology nodes that conventional materials could not reach. CNTs also have excellent electron and hole transport capabilities, and have clean surfaces in terms of not having redundant surface dangling bonds [14].

3 Two-dimensional nanomaterials

One of the usual issues with conventional conductive nanomaterials is their structural brittleness, that makes it difficult to use them in flexible or bendable electronic applications. The inherent properties of two-dimensional (2D) nanomaterials, such as exceptional mechanical flexibility, excellent electrical conductivity, atomic-scale thickness and large specific surface area enable their use in various flexible and bendable devices [1], [15], [16]. The 2D nanomaterials that have shown exceptional results and thus have had large amounts of research done about them are graphene, transition metal dichalcogenides and 2D MXenes. In addition to these, other 2D nanomaterials such as hexagonal boron nitride, black phosphorus and metal-organic frameworks have shown exceptional applicability in certain niche applications.

3.1 Graphene

Graphene is one of carbons allotropes, that has a unique one layered structure, where carbon atoms have attached themselves in a hexagonal, honeycomblike layer as depicted in Figure 3.1 and 3.2 [1], [17]. The carbon atoms are held together by strong sigma bonds in a 0,334 nm thick layer, making it one of the thinnest materials known and giving it the properties of great mechanical flexibility and stiffness with a Young's modulus of around 1 TPa [1], [15].

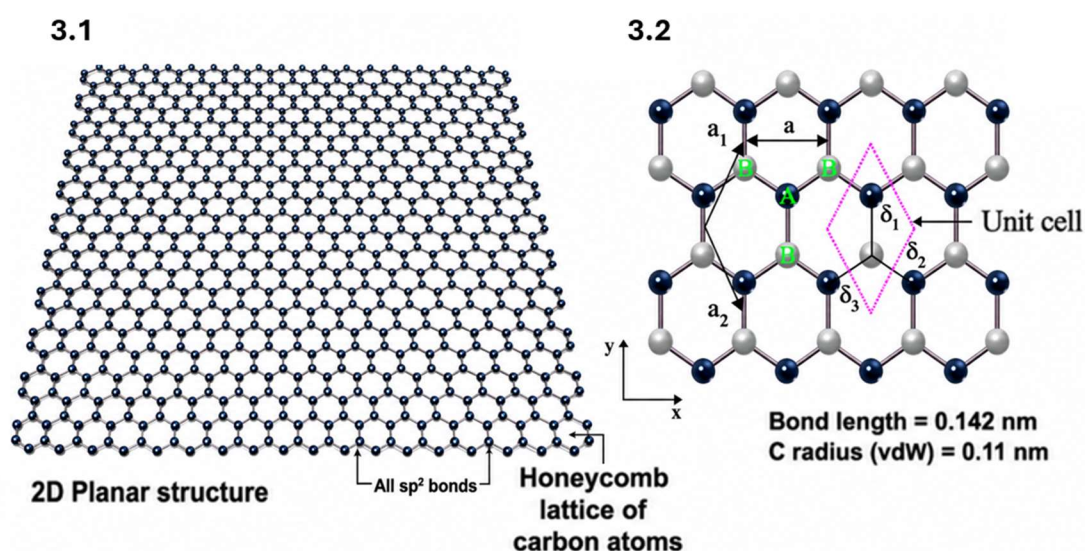


Figure 3. 3.1) Graphene atomic layered structure, 3.2) The honeycomb atomic configuration of single layered graphene, where a structure of one A atom has three neighbouring B atoms, repeating in the lattice. Image reproduced with permission from [18], licensed under CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>).

Due to its unique structure, graphene is also an excellent conductor of electricity. In the structure, each carbon atom uses only 3 of their 4 valence electrons, leaving the fourth to be able to freely move across the lattice. These free electrons behave as charge carriers that are able to travel at high speeds, giving graphene excellent sensitivity in sensor applications [1], [18].

Graphene can be produced using a chemical vapor deposition method (CVD), which allows graphene to be produced in large areas [1], [19]. Despite its advantages, graphene lacks a bandgap which greatly limits its usage in digital devices. However, transistors based on graphene have the advantage of having a low on/off ratio due to graphene's gapless structure, making it ideal for analog devices like sensors. Due to its strengths, graphene has been widely researched in flexible electronics applications as channels, electrodes and sensors [1], [17]. A flexible graphene-based humidity sensor was constructed by using graphene derivatives such as reduced graphene oxide (rGO), to detect and monitor human respiration, sweat and environment humidity [17].

3.2 Transition metal dichalcogenides

Transition metal dichalcogenides (TMDs) are a newly discovered group of 2D nanomaterials, that can have a wide variety of electrical properties depending on the chemical composition and structure. Their structure (depicted in Figure 4) consists of mainly covalent bonds and weak van der Waals forces between other chalcogen layers. TMDs share a chemical structure of MX_2 , in which M represents transition metals like molybdenum (Mo) and tungsten (W), while X denotes chalcogens such as sulphur (S) and tellurium (Te) [20].

The combination of elements that can be used in TMDs, as well as the diverse structure and mono- and multilayer possibility makes TMDs extremely versatile in electronics. TMD properties can span from semiconducting, superconducting and insulating depending on the electronic structure. Therefore, the properties of TMDs can be greatly influenced depending on the component, also assisted by the tunable energy band [20].

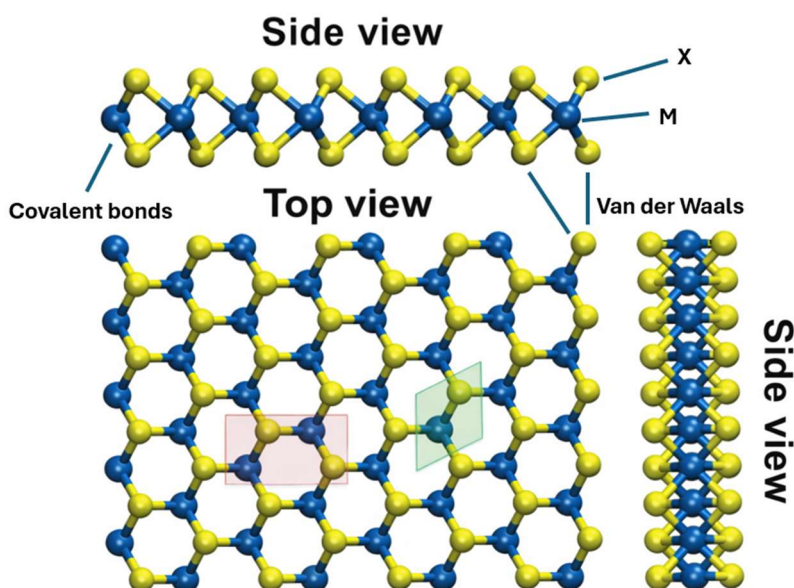


Figure 4. A representation of the TMD atomic structure, where M and X components and the atomic interactions are shown. Image reproduced with permission from [21], licensed under CC BY 3.0 (<https://creativecommons.org/licenses/by/3.0/>).

The thickness of TMDs is also a mechanical strength, that is a great property in flexible electronics. Due to these strengths TMDs have shown great promises in flexible transistors, gas sensors and photodetectors. The ability to fabricate flexible transistors is essential in flexible electronics, for it is a fundamental component in every type of circuit [20]. Current challenges with the utilization of TMDs are that there is still very little research on the fabrication methods and material synthesis, which are both required to fully integrate TMDs into flexible electronics.

3.3 2D MXenes

MAX phases are a group of over 70 compounds, and get their name from their shared general chemical formula of $M_{n+1}AX_n$, where similarly to TMDs, M is a transition metal, A depicts a group 13 and 14 elements, X denotes carbon and/or hydrogen and n represents a number between 1 and 3 [22]. MAX phase structure is illustrated in Figure 5. The M-A and M-X bonds in the MAX phases have very different properties and strength. M-X bonds are a mix of different bond types, such as ionic and covalent bonds, while M-A bonds consist purely of metallic bonds.

The variety of different bonds make the M-X bonds significantly more robust than the M-A bonds, and therefore by synthesizing a material with only M-X bonds could lead to an overall

stronger structure. In 2011 it was discovered that by using chemical exfoliation and selective etching methods, the elimination of the A atomic layer from the MAX phase structure was possible [2], [22], [23]. The removal of the A atomic layer led to the discovery of a family of 2D metal carbides, that were named MXenes, symbolic of the A layer removal [2].

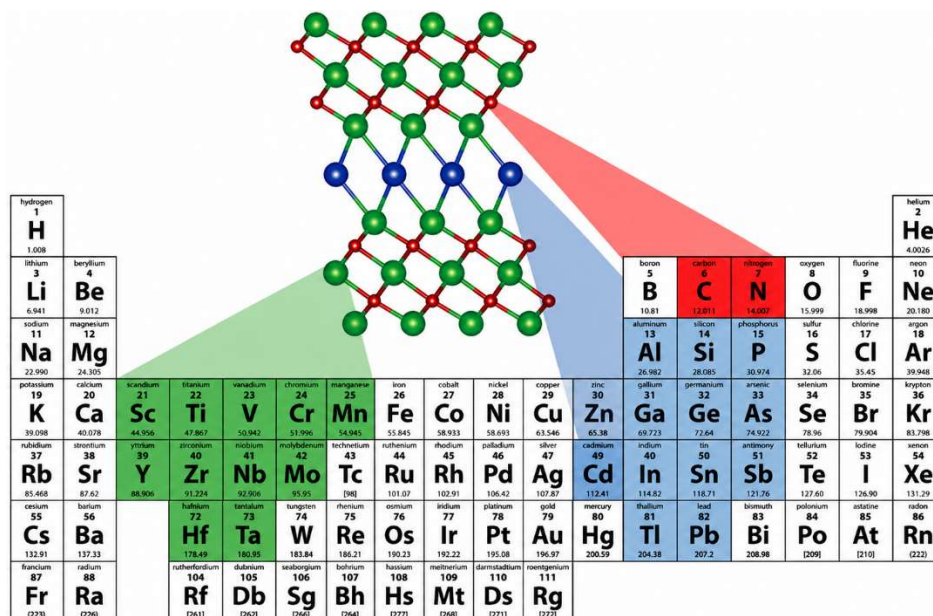


Figure 5. A representation of the elements that form the MAX structure. Reproduced with permission from [22]. Copyright © 2019 Elsevier

MXenes are a group of 2D nanomaterials belonging to transition metal carbides, nitrides and/or carbonitrides. MXenes share a similar common formula with MAX phases ($M_{n+1}X_nT_x$) where $n = 1, 2$ or 3 , M depicts a transition metal, X is a carbon or hydrogen and T denotes negatively charged surface functional groups such as hydroxyl (-OH), oxygen (-O) or fluorine (-F) [6],[7],[23]. After the first MXene was discovered ($Ti_3C_2T_x$), nearly 30 other compounds have been synthesized. On top of the discovered ones, countless more have been theoretically calculated to be possible [22].

In flexible materials manufacturing, MXenes have many desirable properties such as their mechanical fortitude, great electrical conductivity and adequate surface functional groups [2], [23]. The electrically conductive properties of a MXene highly depend on the surface functional groups created during synthesis. Single layered MXenes without any surface functional groups display properties closely associated with metals, whereas MXenes terminated by oxygen exhibit semiconductive properties [2]. MXenes' sensitive, conductive and flexible mechanical properties make it a great base to manufacture sensors with. MXene-based flexible piezoresistive and -electric sensors are perhaps the most promising sensor application. These

piezoresistive sensors work as a convertor of pressure sensations into a resistance signals. They are used in various electronic applications such as displays and electronic skins [24]. A weakness of MXenes is that they are extremely prone to oxidizing to their respective oxides. This process is sped up under humid or damp conditions and warm temperatures. As an example, the before mentioned $\text{Ti}_3\text{C}_2\text{T}_x$ MXene degrades into a $\text{Ti}_3\text{C}_2\text{T}_x / \text{TiO}_2$ complex structure, and finally into TiO_2 particles losing its functionality [2].

3.4 Other 2D nanomaterials

On top of the three prominent 2D nanomaterials discussed before, there are multiple other materials that are being researched to fit the needs of flexible electronics. Some of these 2D nanomaterials are hexagonal boron nitride, black phosphorus and metal-organic frameworks.

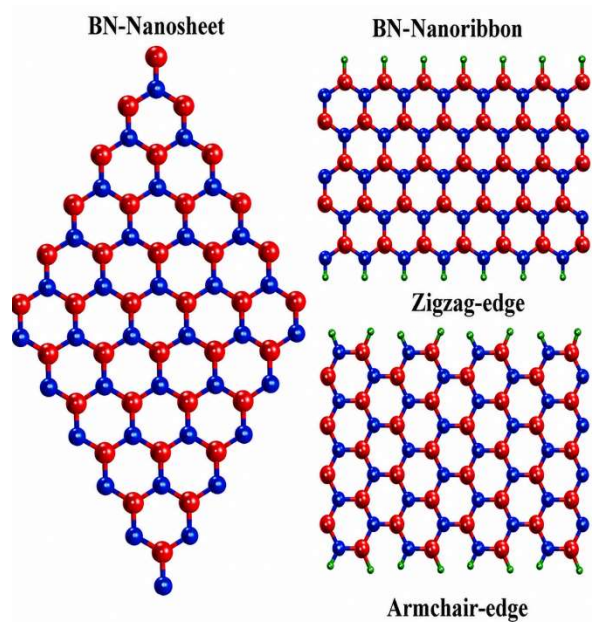
Hexagonal boron nitride (h-BN), nicknamed “white graphene”, is a promising 2D nanomaterial for electronic applications due to its great thermal durability, exceptional thermal conductivity and amazing electrical properties. It has a structure of hexagonally attached boron and nitrogen atoms that are arranged as a sheet as depicted in Figure 6.1. Due to its exceptionally high thermal conductivity, h-BN has shown promise as a thermal interface material, which is a material that is placed in between two component layers to enhance their efficiency under heat [25]. This could enable h-BN to be used in combination with other 2D nanomaterials to create more efficient and structurally stable hybrid structures. However, a known issue with h-BN is its poor cross-plane thermal conductivity. This means that although heat disperses evenly across one plane of the material, its ability to conduct heat onto other layers is not great. This reduces BN’s practical application possibilities considerably [25].

Black phosphorus (BP) is an emerging layered 2D nanomaterial that unlike other phosphorus allotropes, displays great thermodynamic stability. BP has a structure (Figure 6.2) where atoms are bonded with weak van der Waals interactions which gives it the properties of fast ion diffusion and simple intercalation. Another structural benefit of BP is its superb packing density ($2,69 \text{ g cm}^{-3}$) compared to graphene ($1,1 \text{ g cm}^{-3}$). BP has already been used in Li- and Na-ion batteries, supercapacitors, field-effect transistors, gas sensors and some biomedical applications [25].

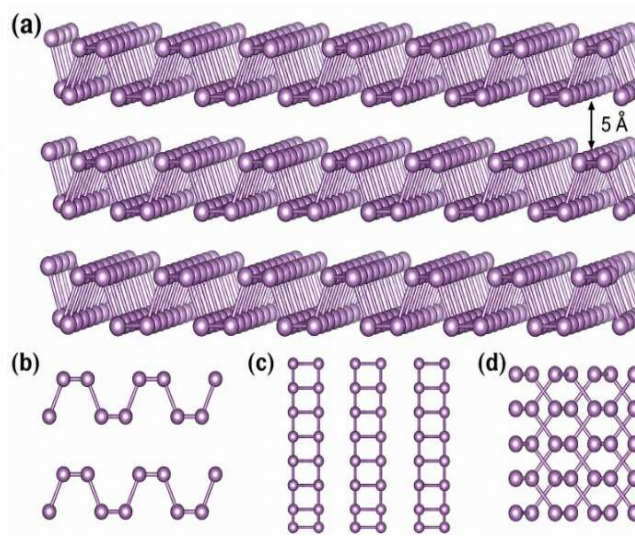
Metal-organic frameworks (MOFs) are a newly discovered chemical sensing material, that has a high porosity and a wide range of possible chemical properties. By themselves MOFs have a distinct weakness in their poor conductivity, that has led to the discovery of 2D conductive

MOFs (2D c-MOFs). 2D c-MOFs do not achieve an electrical conductivity as high as some of the other 2D nanomaterials, but combined with the adjustable porous structure (Figure 6.3) it reaches conductivity that is more than sufficient. Another advantage c-MOFs have, is that their manufacturing process is immensely more efficient than some of the other multi-step processes for other 2D nanomaterials. The efficient synthesis and the high porosity and surface area make 2D c-MOFs a great candidate for sensing applications, namely flexible gas sensors [27].

6.1



6.2



6.3

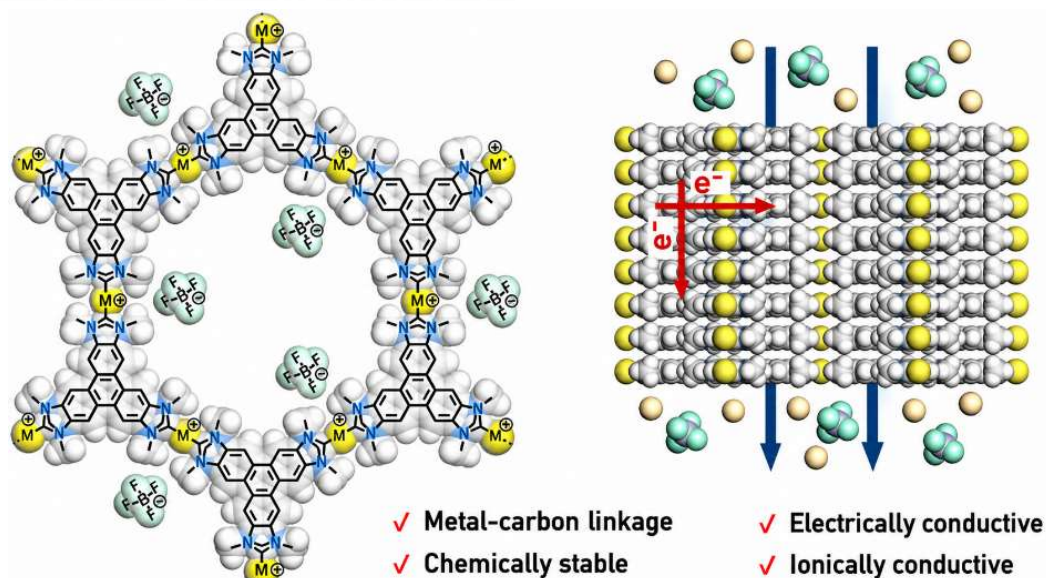


Figure 6. 6.1) Atomic structures of h-BN in three of the most common orientations. Image reproduced with permission from [25]. Copyright © 2025 Elsevier. 6.2) Atomic structure of 2D BP, a) Structure of three-layered phosphorene, b) from the side, c) from below, d) from above. Image reproduced with permission from [26], licensed under CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>). 6.3) The porous and layered structure of c-MOFs, reproduced with permission from [28]. Copyright © 2026 American Chemical Society.

4 Hybrid nanomaterials

By themselves, 1D and 2D nanomaterials excel in both electrically and mechanically challenging applications. Their excellent properties in these fields are the reason why they have shown great promise in the fabrication of flexible electronics. The current issue with these materials is that their intrinsic weaknesses greatly limit what each material can accomplish by itself. 1D nanomaterials, such as AgNWs, have great electrical conductivity, but are disadvantaged by their high junction resistances between wires. As a 2D nanomaterial, graphene has excellent electrical conductivity and great mechanical properties but is extremely difficult and expensive to manufacture. The high cost and complicated synthesis set limitations on what and how graphene can be used in practical applications.

To combat these weaknesses, it is crucial to combine multiple different nanomaterials to complement each other's strengths and weaknesses. Research on 1D/1D, 1D/2D and 2D/2D hybrid nanomaterials has already been conducted to find a combination of materials that would achieve high electrical conductivity, mechanical flexibility and great thermal properties. When it comes to mechanical flexibility, it is also important that the hybrid material does not lose its conductive properties overtime under stretching and bending.

4.1 1D/1D nanomaterials

By themselves, 1D nanomaterials have excellent flexibility, mechanical strength and electrical conductivity. However, the gaps in most 1D nanomaterial networks negatively affect the electrical conductivity of the components they are used in. 1D/1D nanomaterials have shown that these gaps can be filled with another 1D nanomaterial, reducing empty space in the structure and enhancing the conductive properties of the hybrid networks [1]. An example of a 1D/1D nanomaterial is when AgNWs and CNTs are combined into a hybrid nanostructure. Not only does the CNTs improve the AgNWs conductivity, but it also strengthens its structure and greatly reduces resistance under stretching as shown in Figure 7 [1].

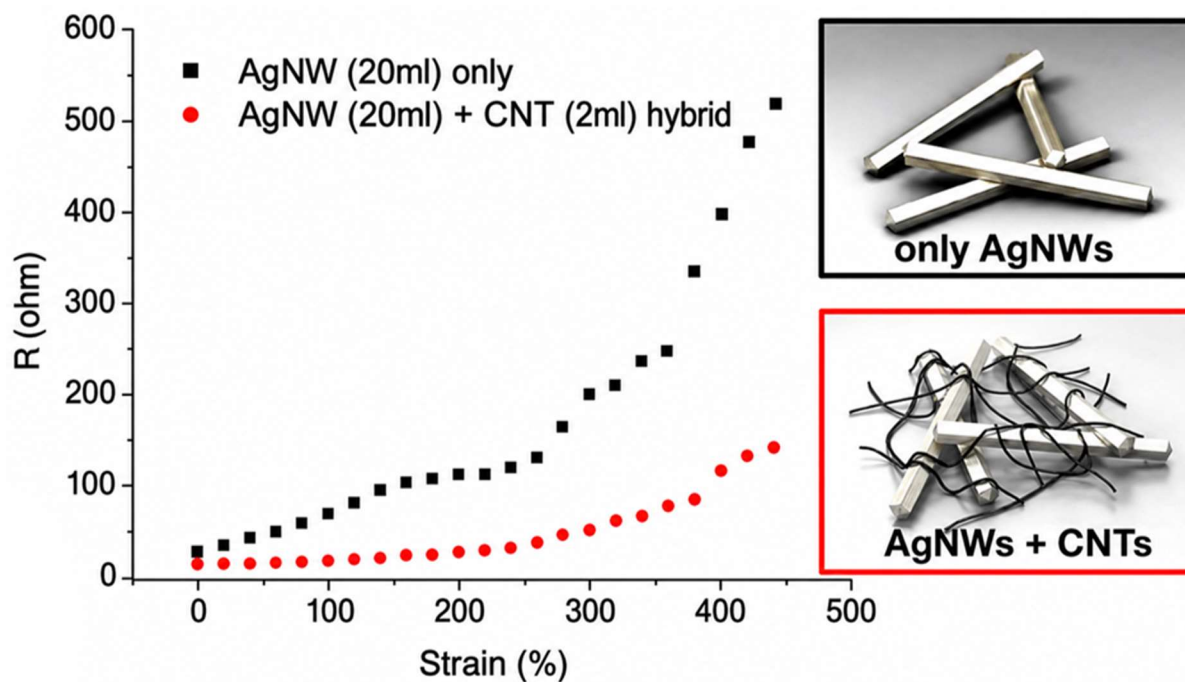


Figure 7. Hybrid AgNW/CNT structure effect on the resistance under load. Image reproduced with permission from [1], licensed under CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>).

Also, by using SWCNTs a hybrid structure where CNTs encased AgNW junctions increasing the bond and ultimately preventing their separation under extreme stretching conditions. The extremely thin structure of SWCNTs also enables the SWCNT-AgNW hybrid's use in optoelectronics. These AgNW-CNT hybrids were used to construct conductors that were able to be used in flexible light-emitting diodes (LEDs) [1].

Another attractive combination of nanomaterials are CNT-MOF hybrids. A notable weakness of CNTs is that they are susceptible to agglomeration and are very difficult to separate with commonly used solvents. As mentioned before, MOFs are known for their poor conductivity and stability but have a high porosity and an easily tunable pore structure. The goal of CNT-MOF hybrids is to attach or grow MOF groups on the surface of the CNT, either by covalent or non-covalent methods as shown in Figure 8. In the covalent method, chemical groups are attached on the surface of the CNT with a covalent bond, that can then spark nucleation of MOFs. The addition of certain functional groups makes the CNT more hydrophilic and thus more practical to work with in certain scenarios. The covalent method does however expose the CNT to oxidation, which decreases conductivity. The non-covalent method avoids impacting the CNT structure, by wrapping molecules around the CNT where they then participate in the forming of MOFs.

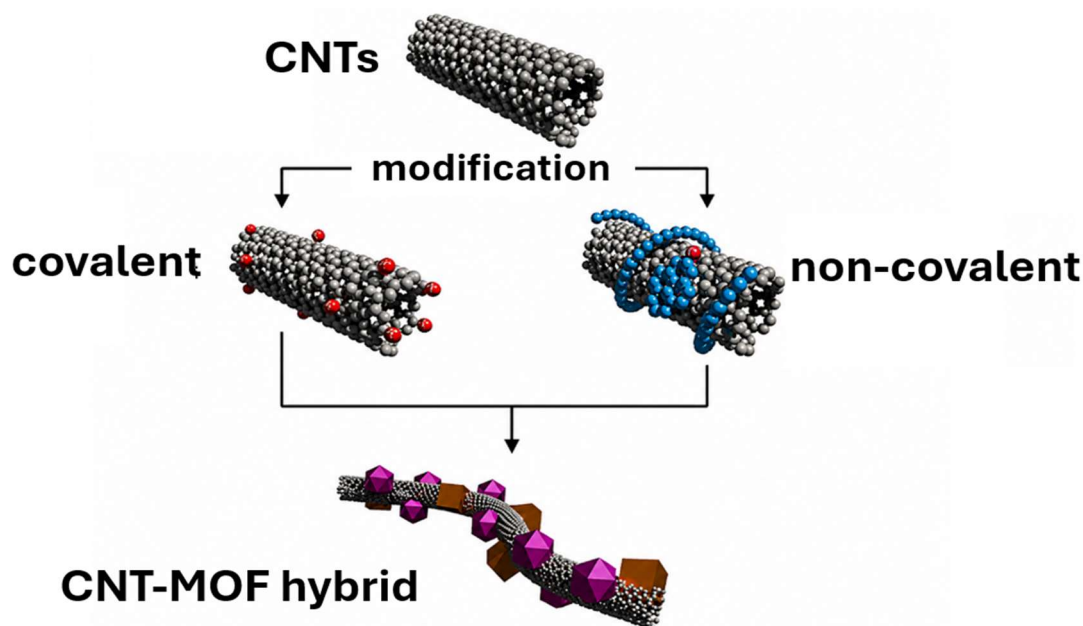


Figure 8. Strategies for the fabrication of CNT-MOF hybrids by covalent and non-covalent methods. Image reproduced with permission from [29], licensed under CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>).

The combination of these structures creates a highly conductive, flexible and porous composite, which has possible applications in water purification, gas sensors and supercapacitors. However, these methods have only been researched with the most common types of MOFs and are still difficult to produce [29]. In this composite, the MOFs are practically not in 1D form, but rather separate nanostructures that are grown onto the surface of the CNT. Nevertheless, it is still a great method of combining nanomaterials to achieve a more functional 1D nanomaterial.

4.2 1D/2D nanomaterials

Despite their strengths, 2D nanomaterials such as graphene have a number of limitations that prevent their use in certain electronic applications. For example, by itself graphene has excellent conductive properties, but there is a limit to how many graphene layers can be layered on top of each other caused by high resistance between its layers [1], [30]. Thus, many 2D nanomaterials have been combined with 1D nanomaterials to compensate for each other's limitations. This problem can be solved by adding a conductive structure, such as CNTs, in between the graphene layers, creating a conductive interface. These graphene-carbon nanotube (GCNT) have shown excellent capabilities in electrochemical energy conversion and storage device applications [30].

Aside from graphene, CNTs have also been combined with other 2D nanomaterials such as MXenes and TMDs. TMDs are known to have limitations in the electronic applications they can be used in, since they are poorly compatible with metal electrodes due to large contact resistance with most metal-based electrodes and poor fracture strain of the metal when in contact with the TMD. The combination of TMDs with other nanomaterials such as CNTs has exhibited lower contact resistances between the materials, while preserving the flexibility and electrical conductivity of both materials. A TMD device was fabricated, where a chemically synthesized TMD (MoS_2) channel was combined with a patterned CNT/ MoS_2 hybrid electrode. The device exhibited excellent electromechanical stability even under considerable bending strain and a superb photoresponsivity compared to conventional metal-based MoS_2 devices [1].

4.3 2D/2D nanomaterials

From the hybrid nanomaterial groups covered in this section, 2D/2D nanomaterials are by far the least researched. Many 2D nanomaterials exhibit similar strengths and weaknesses, thus their combinations would not complement each other too well. Nevertheless, some 2D/2D hybrid structures have been researched, where the two 2D nanomaterials have enough variety in their strengths to be able to create a viable hybrid material.

Perhaps the most lucrative 2D/2D nanomaterial that has been researched is MXene-graphene hybrids. Combining the highly conductive, easily tunable and hydrophilic MXene with the stiff, mechanically strong and conductive graphene would create a unique 2D/2D hybrid structure. One of the ways to approach the synthesis of such hybrid is to layer MXenes with graphene derivatives such as graphene oxide (GO) and rGO [16]. This type of hybrid structure was achieved by using hydrothermal techniques, where MXene and rGO structures were first prepared at 65 °C whereafter the structures went under a freeze-drying process as shown in Figure 9. This resulted in a rGO core-MXene shell hybrid aerogel structure, which exhibited excellent electrochemical properties such as high capacity, heightened cycling life and low capacity decay rate [16].

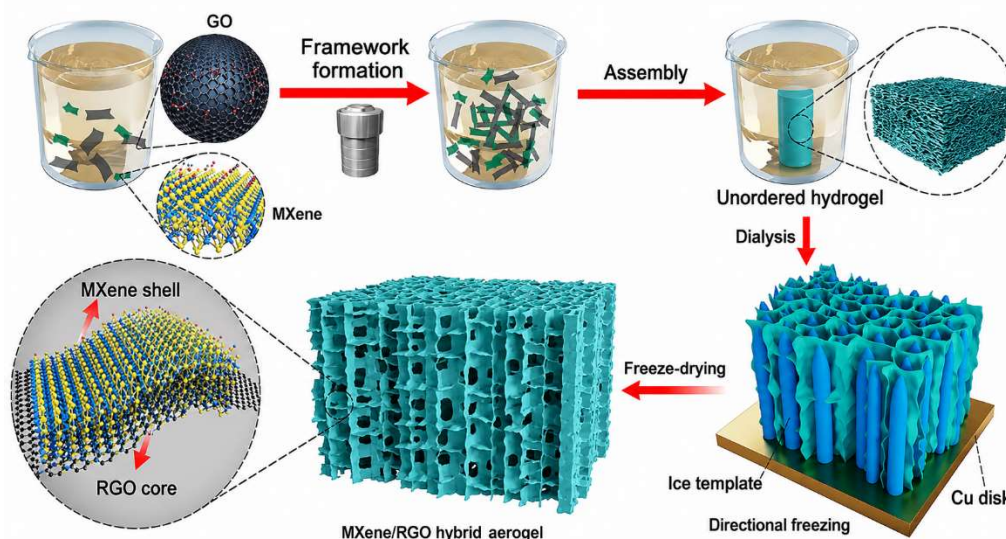


Figure 9. The preparation process of MXene-reduced graphene oxide (rGO) hybrid aerogels. Image reproduced with permission from [16], licensed under CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>).

Another synthesis method used in creating a MXene-graphene composite, was heterogenous self-healing assembly method, which allows the material to heal itself with the use of ambient moisture [15]. This creates a substrateless network where MXene works as the conductor and GO provides functional groups for the moisture activated bond reformation. MXene/GO composites exhibit increased electrochemical properties due to the combined properties of the MXene/graphite layers, and due to the self-healing properties they have potential for wearable electronics and soft robotics applications [15].

5 Hybrid nanomaterial-based flexible electronics

Hybrid nanomaterials have become a trending field of research in electronics manufacturing. The versatility of combined nanomaterials offers a diverse range of excellent electrical and thermal conductivity, mechanical flexibility and high aspect ratios. Hybrid nanomaterials are being utilized in electronics such as field-effect transistors, sensors and optoelectronics. Especially carbon-based nanomaterials such as CNTs and graphene are great candidates as new materials in flexible electronics manufacturing [1].

5.1 Field-effect transistors

Transistors are a key part of nearly every display and sensor device. A variety of different types of transistors are used in flexible electronics. Flexible displays and sensors naturally require flexible components. Thus, the transistors being used in them have to also be flexible [1]. Flexible electrodes for these transistors have been studied extensively, and in these studies, hybrid nanomaterial-based electrodes have proven to be excellent options. A promising candidate for the flexible electrodes has been CNT-graphene hybrid structures [19].

Field-effect transistors (FETs) are one of the most widely used transistor types. FETs can be used as switches, amplifiers and transducers. They consist of a gate-, source- and drain electrode, a semiconductor channel and gate dielectric [1]. FETs have shown to have excellent properties in various applications due to their nanoscale size, high reliability and versatility on how they can be used. FETs are fundamental in the fabrication of communication devices, displays and computers. However, their use as sensors and biosensors in the human health monitoring field has been the most pursued application. The size, sensitivity and multifunctionality of FETs makes them ideal for biomarker detection, making it even possible to detect certain types of cancer [31].

A wide range of different types of nanomaterials have been used in FET synthesis. 1D nanomaterials such as CNTs and silicon nanowires (Si-NWs) have been promising candidates for biosensing FETs. From the types of CNTs, SWCNTs have proven excellent sensitivity compared to the other types of CNTs. 2D nanomaterials have also been used in the synthesis of FETs. Graphene and MXenes have both shown excellent applicability in FETs, due to their thinness, large surface area and electrical conductivity. All of these nanomaterials are excellent in their own rights and have shown great abilities to detect certain types of biomarkers. However, their sensing range is limited to just a few specific markers, and leaves room for

improvement. Hybrid nanomaterials could be a new way to broaden the range of the biosensing FETs and possibly create a FET that would be capable of sensing a multitude of markers simultaneously [31].

5.2 Sensors

Wearable electronics have attracted an increasing amount of attention in the last decade, especially in the health monitoring field. The use of wearable electronics in human health monitoring is especially attractive because it makes disease detection easier and faster, thus improving the lives of many. It has the potential to help prevent certain scenarios and makes it easier for individuals to know what is happening in their bodies. For wearable electronics to be able to detect changes in human bodies or the environment, flexible and wearable sensors must be fabricated [1] [23].

Conventional approaches in flexible sensor fabrication still require betterments in signal detection sensitivity, conductive consistency and sensing range. Thus, flexible sensors using hybrid nanomaterial structures have been researched extensively. Hybrid nanomaterial structures such as CNT-graphene have shown great performance as pressure sensors. CNT-based sensors by themselves have shown to have high flexibility but poor sensitivity, while graphene-based sensors have exhibited excellent sensitivity but a limited range of sensing [19]. CNT-graphene are especially promising in physical sensors such as pressure and strain sensors. Much research has been conducted on MWCNT-graphene pressure sensors, because of their combined strengths being ideal for flexible sensor manufacturing. Combined, they achieve high sensitivity, excellent flexibility and low detection limit, which are great for applications like artificial skins. Durable and flexible electronic textile-based strain sensors have also been researched using CNT-graphene structures. These e-textile-based strain sensors were achieved using CNT-rGO hybrid material in a non-woven fabric. Despite their advantages, CNT-graphene based sensors still face the same challenges in cost-effectiveness and fabrication complexity. Researchers have tried methods of synthesizing these sensors, such as direct laser writing and 3D-spraying methods, that have shown promising results in efficient fabrication [19].

CNTs have also been combined with MXenes in the fabrication of 64-channel flexible bioelectrical signal sensor arrays. This hybrid structure-based sensor array showed excellent stability and maintained steady signal acquisition under complex environments. This concept

was further developed in the form of a combination of silver nanoparticles, CNTs and graphene nanosheets, creating a 0D/1D/2D hybrid nanomaterial. This structure combined the ascended surface activity of silver particles, the superb electrical conductivity and mechanical flexibility of CNTs and the mechanical stability of graphene. The bioelectrical signal sensor made using this structure demonstrated great stability during dynamic strain and maintained superb performance under multiple stretching cycles [32].

2D/2D hybrid nanomaterials have also shown to have some applications in sensor fabrication. Especially MXene-graphene hybrids have shown great promise in biosensing applications. Graphene-based materials have been used extensively in biomedical applications, so by creating a hybrid material based on graphene could lead to excellent new materials. Researchers were able to fabricate FET-based biosensors using MXene-graphene hybrid structures to be able to detect influenza and coronavirus with outstanding chemical sensitivity. 3D porous MXene-graphene composite films were also used to produce biosensors used as glucose detectors. By controlling the ratio of MXene and graphene nanosheets, the size of the internal porous structures could be customized to positively affect glucose biosensing efficiency [16].

A notable challenge with these carbon-based nanomaterials and sensors, is that there is still very little data on how human bodies react to these materials in the long term. The toxicological effect of graphene must be studied extensively, to understand how it reacts to human cells and tissues. Materials that inevitably end up on the skin or more so in the human body cannot have any short- or long-term consequences for their user, nor can they lose their properties in those conditions. If they were to lose their properties, they would have to be replaced in certain intervals, which is not a sustainable practice for sensors in the human body [16].

5.3 Energy devices

Due to the increasing interest in wearable electronics and sensors, a need for flexible energy storage devices has also emerged. Conventional batteries such as lithium-based batteries have under extensive research shown to have an increasing risk for exploding under mechanical deformation, thus are not suitable for these flexible applications [1]. In addition to the flexibility requirement, these energy devices must also be able to be easily integrated into wearable electronics. Flexible wire-shaped energy storage devices have been fabricated using N-doped CNT yarns / MXene hybrid structure [33]. This system was mainly fabricated for the possibility of inconspicuous integration into smart textiles. This storage device showed a great potential

window of 2 V and exhibited excellent mechanical stability even after 5000 test cycles. The electrophoresis method used in this device can be easily upscaled to mass production [33].

In addition to flexible energy storage devices, the demand for flexible supercapacitors is also on the rise. Supercapacitors are characterized into groups depending on the mechanism they use to store charge. They are divided into electrical double-layered capacitors (EDLCs), pseudocapacitors and hybrid supercapacitors [1], [34]. EDLCs utilize a non-faradaic process to store charge and are mostly fabricated from carbon-based materials, while pseudocapacitors utilize a faradaic process and are usually based on transition metal oxides and transition metal sulphides. Hybrid supercapacitors are therefore a combination of the two mentioned before, involving both faradaic and non-faradaic processes, and can be fabricated from a combination of the materials mentioned before [34]. 2D nanomaterials such as graphene and TMDs have been researched as the active material in the supercapacitors as they have extremely large active-surface area and versatile electronical properties. Especially TMDs like molybdenum disulfide (MoS_2) have been attractive as a catalyst and electrocatalyst for hydrogen evolution reactions. A challenge with MoS_2 is that its energy storage capacity is greatly limited as it is a semiconductive material. Thus, using hybrid nanomaterial composites such as rGO- MoS_2 has proven to be a great alternative [34].

6 Conclusions

Hybrid nanomaterials represent a multifunctional solution for the field of flexible and stretchable electronics, acting as strong candidates to replace the inherently limited conventional brittle materials like gold, copper, and indium tin oxide. This thesis has found that while individual 1D nanomaterials, such as silver nanowires and carbon nanotubes, and 2D nanomaterials, such as graphene and MXenes, possess exceptional electrical and mechanical properties, their practical application is often limited by specific weaknesses like high junction resistance, oxidation susceptibility or high fabrication costs [1].

The core finding of this thesis is that hybrid nanostructures including 1D/1D, 1D/2D, and 2D/2D combinations effectively mitigate these weaknesses. As an example, combining carbon nanotubes with silver nanowires reduces junction resistance and enhances structural integrity under mechanical load. Similarly, 1D/2D hybrids like graphene-carbon nanotubes overcome interlayer resistance, proving highly effective for electrochemical energy conversion and storage. These materials have enabled significant advancements in field-effect transistors, flexible pressure, strain and gas sensors, supercapacitors and stable energy devices all suitable for human health surveillance, biosensing and integration into smart textiles and wearable electronics.

Despite the progress made in the development and research of flexible hybrid materials, many critical challenges remain before these materials can transition from laboratory prototypes to scalable commercial solutions. Long-term safety and toxicological research must be conducted on materials that are to be in contact with the human body. Especially data on carbon-based materials has to be gathered to get a deep understanding on the long-term effects they have and how they react with human tissue. Many of the methods used in the fabrication of these hybrids are also not sustainable, due to their subtractive nature. Research on additive manufacturing methods would reduce the amount of waste generated from flexible device synthesis [9].

In conclusion, the integration of diverse nanomaterials into hybrid structures provides a diverse platform for the next generation of multifunctional, durable, and flexible electronics. By tailoring material combinations, it is possible to achieve the high conductivity, mechanical robustness, and sensitivity required for the future of wearable electronics, sensors, energy devices and much more.

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