Recent Advances of Graphene and Related Materials in Artificial Intelligence

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Biological brains perform real-time processing of unstructured data with ultralow energy consumption and represent the most efficient computing systems. Emulating the working principles of biological brains brings a revolutionary breakthrough in artificial intelligence (AI), generating two kinds of imitation approaches including machine learning (ML) and neuromorphic computing (NC) with artificial neurons/synapses. Herein this review, a general description of ML methods, the concept, and principle of artificial synapses (ASs) in NC derived from biological synapses, and their relationships, is provided. Graphene, one of the most representative 2D materials, arouses considerable attention due to its unique structure and properties. The application of ML in properties prediction (electronic properties, mechanical properties, thermal properties, cytotoxicity), structure recognition (atomic structure, microscopic dimensions/shapes), inverse design (composition, structure), and task recognition (chemical recognition, motion recognition, 3D imaging) of graphene and its derivatives and composites are summarized, and corresponding methods are discussed in the case studies. Recent progress in the development and application of graphene-based ASs (synaptic transistors and memristors) is briefly introduced, where graphene-based materials serve as the channel materials of synaptic transistors, the memristive materials, or back electrodes of synaptic memristors. Finally, the main challenges and prospects of graphene-incorporated ML and ASs are presented.

1. Introduction

Since the inception of the computer era, researchers have been striving to use computers to acquire new declarative knowledge, develop motor and cognitive skills, and discover new facts and theories. This is the origin of developing artificial intelligence (AI), which aims to make a machine capable of human-like intelligence. AI uses computers or machines to emulate the problem-solving and decision-making capacities of human intelligence. The von Neumann computers are ideal platforms for solving structured problems. However, the information processing relies on the frequent data transmission between processors and memory units, which inevitably limits the computational efficiency and scalability. In addition, the energy consumption increases dramatically with the increasing computing speed. The problem gets worse in dealing with unstructured data (e.g., video, image, voice recognition, information retrieval). In contrast, the human brain is a highly efficient biological computing system that can perform real-time processing of unstructured data with ultralow energy consumption. Emulating the biological brain is the only way to reach a higher level of AI. There are two ways to imitate the functions of a biological brain, including machine learning (ML) with software and neuromorphic computing (NC) with hardware.[1]

The discovery of graphene in 2004 by A. K. Geim et al. has raised an extraordinary amount of interest of 2D materials, leading to a revolutionary development of many 2D layered materials such as metal chalcogenides, transition metal oxides, and other 2D compounds. Graphene is the most widely studied one among 2D materials, showing the highest stability, excellent electron mobility, and a mature preparation process of wafer-sized single crystals. Graphene has a 2D structure with sp²-bonded carbon atoms arranged in a honeycomb or hexagonal lattice (Figure 1). Each carbon atom has 4 valence electrons: 3 electrons form plane sp² hybrid orbitals, and the remaining one is used to form a large π bond. Graphene is a building block of carbon materials such as graphite (3D), carbon nanotubes (1D), and fullerene (0D). The unique atomic structure endows graphene with excellent properties such as zero-gap band structure, high visible light transmittance (97.7%), high electron mobility (10^6 cm² V⁻¹ s⁻¹), high electrical conductivity (10⁶ S cm⁻¹), high thermal conductivity (5000 W m⁻¹ K⁻¹), excellent mechanical flexibility, high Young’s modulus (1 TPa), and large specific surface area (2600 m² g⁻¹). Compared with graphene, graphene oxide (GO) containing both sp² and sp³ domains possesses many oxygen-containing functional groups (e.g., hydroxyl, epoxy, carboxyl, and carbonyl groups), which can significantly affect its physical and chemical properties. GO can be reduced to
graphene (reduced GO, RGO) by a reducing agent but remaining inevitable structural defects. Many graphene-based devices have been applied in optoelectronics, surface catalysis, antifriction, and coatings.\textsuperscript{[2]}

Recently, the application of graphene in AI has aroused considerable attention. On the one hand, graphene has been widely used to build wearable electronic devices (e.g., sensors) to mimic the capabilities of human senses controlled by the human brain such as touch, smell, taste, and hearing. Graphene sensors are combined with ML techniques to implement task recognition, where graphene sensors detect the external environments and generate output signals, while ML techniques employ the information to establish the relationship between the known environments (e.g., liquid, gas, motion, voice, and image) and output signals (usually electrical signals). Associations constructed from the inputs and outputs of the training set by ML models are suitable for the detection of unfamiliar environments.\textsuperscript{[3]}

In addition, ML methods are suitable for properties predictions, structure recognition, and inverse design of graphene.\textsuperscript{[4]}

On the other hand, graphene is recently used to construct artificial synapses (ASs) in neuromorphic systems. Graphene has been used to fabricate synaptic transistors and memristors, emulating the synaptic plasticity in the biological brain such as potentiative and depressive plasticity, and modulatable plasticity. When serving as the channel material in synaptic transistors, the characteristic of zero-bandgap of graphene allows the gate voltage to tune the conduction state of graphene. The transition between electron- and hole-dominated conductions allows both behaviors of excitatory and inhibitory synapses to be realized in a single graphene transistor.\textsuperscript{[5]} When acting as the memristive material or back electrode in synaptic memristors, graphene-based memristive materials can perform memristive function through various mechanisms such as defect migration, vacancy migration, filament formation, and charge trapping,\textsuperscript{[6]} while graphene-based back electrodes can reduce the running current, increase the on/off ratio, and lower the energy consumption of memristors due to their high out-off-plane contact resistance.\textsuperscript{[7]}

Memristors possess the current–voltage characteristics with a closed hysteresis loop pinched in the origin, showing an intrinsic nonvolatile synaptic plasticity.

This article reviews and discusses the latest research and development of graphene and its derivatives in AI, including ML techniques and ASs devices (Figure 1). The content is divided into four parts: 1) Basic concepts and principles of graphene materials, AI, ML, and ASs; 2) application of ML in properties prediction, structure recognition, inverse design, and task recognition of graphene materials; 3) application of graphene-based transistors and memristors in ASs; and 4) Summary and outlook of graphene-incorporated AI.

### 2. Artificial Intelligence

#### 2.1. Machine Learning

In the fields of drug discovery and materials research, ML approaches have been applied to guide time-consuming experiments and theoretical calculations. Generally, the quantitative structure–property relationship (QSPR) between structure descriptors and target properties is generated by ML models to identify potential target structures quickly yet cheaply with required properties from millions of candidates. Different types of ML methods used for QSPR have their pros and cons.\textsuperscript{[8]}

Linear regression attempts to model the relationship between two variables by fitting a linear equation. Kernel ridge regression (KRR) learns a nonlinear function by employing the kernel trick. Linear discrimination analysis (LDA) is a classifier that employs a linear equation to maximize the between-class distance and minimize the within-class distance. Support vector machine (SVM) classifier separates different categories by mapping points in space to maximize the margin between different classes of points. SVM regressor (or support vector regression, SVR) is to approximate the best value within a given margin. Decision trees (DT) classifier or regressor creates a model by learning simple decision rules inferred from the data features. Random forest (RF) classifier or regressor is an ensemble method operating by constructing multiple DTs as base learners. The k-nearest neighbors (kNN) is an unsupervised algorithm for classification and regression, operating by counting the class of k nearest neighbors in the feature space. Clustering is an unsupervised algorithm for dividing data points into several groups such that similar data points are grouped. The artificial neural network (ANN) is a computational network emulating the biological brain, composed of three basic layers (one input layer, one or two hidden layers, and one output layer).\textsuperscript{[9a]}

Unfortunately, most traditional QSPR studies use a single modeling approach to develop a single model based on one type of descriptor, only studying part of the chemical or biological diversity of a dataset. In addition, in the current big data era along with the rapid growth of data scale, diversity, and uncertainty, traditional QSPR approaches derived from ML models are not suitable. Consequently, ANN is further developed to introduce some models such as multilayer perceptron (MLP) or deep neural network (DNN), convolutional neural network (CNN), recurrent neural network (RNN), and deep belief network (DBN), generating a new branch (deep learning, DL) of ML.
The DL models use large numbers of hidden layers, and each layer comprises hundreds of neurons. The DNN contains multiple hidden layers which are fully connected. The CNN contains several convolution layers and subsampling layers. The convolution layer has a set of filters (or kernels) that have a small receptive field and learnable parameters. The subsampling layer is used to reduce the size of feature maps. The RNN allows the connection among neurons in the same hidden layer to form a directed cycle, suitable for time-dependent tasks. The DBN is a probabilistic generative model, composing of stacked modules of restricted Boltzmann machines (RBMs). These DL models can transform the representations of features from low- to high-level features without the manual selection of descriptors, benefiting to employ raw high-dimensional data to build the QSPR.[8]

2.2. Artificial Synapses

2.2.1. Software and Hardware for ANN

A great number of neurons ($\approx 10^{11}$) and synapses ($\approx 10^7$) are responsible for the information processing in the human brains, where neurons receive, process, and transmit the information as electrical and chemical signals, while synapses store and process information simultaneously. There are two main approaches to simulate the neural network in the human brain and thus implement brain-like computing. One is software simulation which performs calculations in von Neumann computers using ANN models with linked nodes called artificial neurons. The node–edge–node structures of the ANN models correspond to the biological axon–synapse–dendrite architectures, where the weights of the connections determine the node-to-node impact. The purpose of training an ANN model is to optimize the weights. DNN composed of a large number of neuron layers is the best one to address real applications today (Figure 3a). Recently, spiking neural network (SNN) has been rapidly explored, because the discrete spikes are closer to biological neural systems than the static and continuous-value activation used in the conventional DNN. The other is hardware implementation namely neuromorphic systems that construct ANN devices using electronic devices. The general paradigm in neuromorphic systems is to take inspiration from the biological brains to build circuits composed of artificial neurons interconnected by ASs. Therefore, artificial neurons and ASs are building blocks of neuromorphic systems. As synapses connect neurons and perform computing and learning, current research is mainly focused on ASs rather than artificial neurons. Synaptic transistors and memristors are typical ASs used for building a crossbar array to connect two layers of neurons and store weight values (Figure 3b).[1]

2.2.2. Biological Synapses

A synapse plays an important role in the signal transmission (chemical or electrical signals) between two neurons. The comparison of synaptic transmissions between chemical and electrical synapses is shown in Figure 4a. Since electrical synapses are not involved in memory and learning, only chemical synapses found in humans and other vertebrates are the major research objects. All of the biological synapses mentioned later are chemical synapses. The information transmission and processing in synapses are divided into several steps: 1) upon the triggering of action potentials, the voltage-gated calcium...
channels (VGCCs) is opened; 2) synaptic vesicles breakup and neurotransmitters are released, in a probabilistic manner; and 3) ionotropic and metabotropic receptors detect and translate the presynaptic message (neurotransmitters) into various postsynaptic events, including membrane potential, biochemical cascades, and gene expression, to amplify the presynaptic signal. This is the basic information transfer process between two neurons. The release of exhibiting and prohibiting neurotransmitters results in the excitatory postsynaptic current (EPSC) and inhibitory postsynaptic current (IPSC), respectively. The summation of postsynaptic current (PSC) in the postsynaptic neuron determines whether the postsynaptic neuron triggers the action potential or not.[9]

The changes in connection strength (synaptic weight) between neurons can be attributed to synaptic plasticity (potentiation and depression), which is classified as short-term plasticity and long-term plasticity. Short-term plasticity such as short-term potentiation (STP) and short-term depression (STD) occurs from milliseconds to minutes, relevant to the physiological basis for computational functions in the neural network (NN). While long-term plasticity such as long-term potentiation (LTP) and long-term depression (LTD) lasts several hours or longer, relating to learning or memory in the neural network. With the increasing intracellular centration of Ca\textsuperscript{2+}, STP is triggered to facilitate the release of neurotransmitters. Once the concentration reaches a threshold, LTP is triggered to enhance the permeability of primary α-amino-3-hydroxy-5-methyl-4-isoxazolepropionic acid (AMPA) receptors or recruit new AMPA receptors in the postsynaptic membrane, and new proteins are generated to form new synapses (Figure 4b). The long-term and short-term plasticity are reflected by the corresponding PSC (Figure 4c). In addition, the synaptic weight is not only dependent on the time interval between the presynaptic and postsynaptic spikes but also related to the frequency of the individual spikes, corresponding to the spike-timing-dependent plasticity (STDP) and spiking-rate-dependent plasticity (SRDP), respectively. The intensity of the synaptic weight decreases with increasing time interval and decreasing frequency and that occurrence of the pre-spike before or after the post-spike induces either excitatory or inhibitory synaptic behavior, respectively.[9b,10]

2.2.3. Artificial Synapses

Similarly, ASs electronic devices are sensitive to multiple stimuli, thus sensing and processing signals efficiently. When subjecting to different stimuli, such as electric field, magnetic field, light, pressure, and temperature, ASs can operate by various mechanisms to imitate the plasticity in the biological synapse. First, the electric fields can trigger ion migration, electron/hole transfer, phase changes, magnetic domain wall motion, and ferroelectric polarization. Resistive switching (RS) elements typically operate under electrical stimuli to perform binary resistance transitions between the low-resistance state (LRS) and high-resistance state (HRS). In some cases, intermediate-resistance states (IRSs) may appear, involving multiple-resistance states with an improved memory window or enhanced logic states for multilevel data storage. Second, light illumination can cause charge-carrier transport and chemical reactions. The resistance transitions of ASs are regulated by photogenerated charge carriers, which are trapped in specific functional layers, or altered by photoinduced reversible chemical reactions in the channel materials. The devices have a memory effect that is affected by the duration and frequency of the light pulse. Third, based on the piezoelectric effect which converts mechanical stimuli into the electrical signal, piezoresistive switches can be triggered by mechanical forces to perform resistance transitions between the LRS and HRS. Therefore, the changes in connection strength (synaptic weight) are reflected by the corresponding PSC (Figure 4c).
into electric signals, electric signal outputs generated by ASs vary with the external force. Similarly, under the stimulus of a magnetic field, ASs operate through the magnetoelectric coupling effect which converts magnetic signals into electric signals. At last, based on the thermos-electric effect, a direct conversion of electric signals from temperature difference, the output of the ASs is affected by thermal stimuli. All of the changes in electric signals mentioned before are related to the change of synaptic weight of ASs, capable of realizing synaptic plasticity such as STP, STD, LTP, LTD, STDP, and SRDP. The selection of functional materials that respond to different stimuli is the key to fabricating ASs.

ASs have several fundamental metrics, including energy consumption, state retention times, switching speed, cycling endurance, integration scale, and device stability. In detail, 1) the energy consumption should be less than several pJ per spike level, especially when scaling up the device toward the capacity of the human brain; 2) the state retention time can vary from $10^3$ to $10^6$ s, and longer retention times are generally favored; 3) the switching speeds should be less than 1 μs; 4) the cycling endurance should be more than $10^6$ cycles; 5) the device size should be less than 1 μm$^2$; and 6) the device can operate at elevated temperatures of up to 400 °C.

3. ML-Based Prediction and Recognition

ML methods are efficient to build the QSPR of materials. The resulting QSPR not only allows rapid and accurate properties prediction and structure recognition but also benefits the inverse design of graphene and its derivatives. In addition, the ML technique has been widely used to assist the task recognition of graphene-based sensors.

3.1. Properties Prediction

3.1.1. Accelerate Theoretical Calculation

Density-functional theory (DFT) calculations have been extensively used to study the properties of graphene materials with high accuracy and reproducibility. But the computational cost of DFT calculations scales fast with the number of atoms, and these calculations are exceedingly expensive for large systems at finite temperatures. The high computational resource consumption can be attributed to the full calculation of potential energy surface (PES) or the solution of the Kohn–Sham (KS) equation. The ML technique can learn the input–output behavior of the KS equation and predict the electronic density of states (DOS) and charge density, bypassing the KS equation to be a fast and accurate DFT emulator. The electronic DOS together with the charge density can be used to predict various material properties, outperforming DFT in computational time by several orders of magnitude while preserving chemical accuracy. R. Ramprasad et al. applied this approach to study a diverse set of carbon allotropes spanning a large configurational and phase space. From another perspective, ML models can be trained to predict material properties with only a few data obtained by DFT calculations. The association between structural information (e.g., topological, geometrical, or electronic features) and material properties, namely QSPR, is created directly by ML models. The resulting QSPR allows rapid and accurate estimation of material properties from material structures. With the aid of a few DFT calculations and geometrical estimators as the training dataset, L. Qiao et al. demonstrated that the well-trained ML models can accurately predict the mechanical properties of graphene.

Molecular dynamic (MD) is another atomistic-based simulation to explore the material properties, whose computational cost scales linearly with the number of atoms. Empirical interatomic potentials have long provided an indispensable tool in facilitating MD studies. However, the prediction accuracy, reproducibility, and transferability of empirical potentials are still under concern, and the existing interatomic potentials are not available for a majority of novel materials. On the one hand, ML methodologies have been used to develop interatomic potentials such as neural network potential, Gaussian approximation potentials (GAP), spectral neighbor analysis potential, and moment tensor potentials with arbitrarily high accuracy. The machine-learning interatomic potentials (MLIPs) are a powerful assistant to MD studies. A. Michaelides et al. developed an ML model to obtain a faithful representation of the DFT-PES, constructing an accurate interatomic potential (GAP) for graphene. The GAP can quantitatively predict the lattice parameter, thermal expansion coefficient, and phonon properties of graphene with only a marginal compromise on accuracy. The computational cost of the MLIPs-based MD calculations is orders of magnitude lower than that of traditional calculations which invoke electronic structure methods. Therefore, this method would be beneficial to study dynamic cases. In addition, MLIPs can be employed to conduct first-principles multiscale modeling in which ab initio level accuracy can be hierarchically used to explore the properties of macroscopic systems. As shown in Figure 5, ab initio molecular dynamics (AIMD) simulations are first conducted to prepare a dataset for ML training. MLIPs are then developed for the subsequent MD calculations. After that, MLIP-based MD calculations are conducted to evaluate the material properties. Finally, the material properties of macroscopic structures can be examined using finite element modeling (FEM). After evaluating the mechanical properties and thermal conductivity of graphene and graphene/borophene composites using MLIPs-based MD simulations, A. V. Shapeev et al. conducted the continuum FEM to study the stress–strain responses and effective thermal transport of graphene/borophene heterostructures at the macroscopic scale. This method is attractive to develop fully automated platforms to design, optimize, and explore material properties and nanostructures at continuum level with atomic accuracy.

On the other hand, the MD simulation combined with ML strategy presents as an efficient and quick method to construct the QSPR and predict the properties of materials. J. Zhang et al. employed high-throughput computations (HTC) methods to conduct MD calculations as the training dataset of ML models. Through constructing the association between structures (or external environments) and properties, the well-trained ML model is applied to study the influence of external environments or material structures on the mechanical properties of graphene.
3.1.2. Electronic Properties

Electronic properties are essential to understand the complex structure–function relationships at the nanoscale, especially for 2D materials. Through the construction of QSPR between structural features and electronic properties, the DFT calculation combined with ML approach allows accurate properties predictions to be made at a fraction of the computational cost of high-level DFT calculations. A. S. Barnard et al. trained ML models to correlate graph topological fingerprints or structural features to the electronic properties of graphene with different shape, size, and edge conformations in quantitative terms. The ML models can predict the Fermi level energy and bandgap of graphene with an accuracy of about 90%.[17]

Introduction of defects at atomic or nanoscales (e.g., doping) in graphene can manipulate the electronic properties. The properties of doped graphene depend on types of dopants, doping concentrations, and dopants configurations, which may exhibit billions of possibilities. Data-driven ML starts to stimulate great interest to build the sophisticated QSPR. J. Lin et al. conceived the CNN to predict the bandgaps of hybridized (B–N dopant) graphene with any given configurations. Bandgaps of doped graphene with arbitrary concentrations and configurations were calculated by DFT, serving as the training and testing datasets. The 2D matrices were defined as material descriptors to quantitatively capture the features of different configurational states (Figure 6a). The QSPR of structures and bandgaps was built by a well-trained CNN possessing prediction accuracy higher than 90% (Figure 6b). This method stimulated interest in applying ML models for the configurational design of materials.[13a]

3.1.3. Mechanical Properties

The excellent thermal, electronic, optical, and mechanical properties of graphene and its derivatives make them applicable to various applications such as field-effect transistors (FET), supercapacitors, sensors, and optical devices. For example, the mechanical responses of graphene and its composites are vital for their applications. MLIPs-based MD simulation has been regarded as a high-precision and time-saving method to explore their mechanical properties. A. V. Shapeev et al. used MLIPs-based MD calculations to simulate the mechanical response of graphene. Six strained structures calculated by AIMD are considered to construct the moment tensor potentials. The stress-response of graphene calculated by the MLIPs-based calculation excellently reproduces the initial linear part of the stress–strain relation, outperforming that based on classical interatomic potentials. Moreover, the calculation reveals the brittle nature of graphene which is not available from DFT-based calculations. This work also employed the FEM to study the stress–strain responses of graphene-based composites at the macroscopic scale. Differently, J. Wang et al. combined common MD simulations and ML techniques to construct the QSPR to predict the stress–strain response of graphene-reinforced metal matrix nanocomposites. The result was then used to modify the existing Halpin–Tsai model. This method is generic and applicable for other composites reinforced by 2D nanofillers.[18]
The impact factors have an obvious influence on the mechanical properties of graphene, including internal factors (e.g., defects, dopants, chirality, and edge effect) and external factors (e.g., strain rate, system temperature). Accelerated studies about the influence of multiple factors on the mechanical properties of graphene materials can be realized by ML methods. J. Zhang et al. combined ML algorithms (e.g., kNN, SVM, DTs, ANN) with MD simulations to study the effects of system temperature, strain rate, single vacancy defect, and chiral direction on the fracture strain, fracture strength and Young’s modulus of graphene. With QSPR, the well-trained model can predict the mechanical properties of graphene at known system temperature, strain rate, single vacancy defect, and chiral direction.

Another fundamental challenge in technological applications of graphene and its composites is the fracture mechanism, where the crack growth behavior is valuable toward the material design at the nanoscale. A. Tabarraei et al. proposed an ML model comprising a CNN and a bidirectional recurrent neural network (Bi-RNN) to predict the fracture evolution in polycrystalline graphene sheets (Figure 7a). The training dataset is constructed by conducting tensile loading MD simulations on 700 different polycrystalline graphene sheets. The input image represented the microstructure of graphene sheets, while the output image represents the fully developed cracks of graphene sheets. The input–output correlation (QSPR) was then established by the well-trained ML model, predicting the crack path in graphene sheets with high accuracy. As shown in Figure 7b, crack growth obtained from the ML model agrees well with the MD simulations. The ML method is suitable for image-based matrices scrubbed of any discrete atomic information, thus it is not limited to particular material scales and may be a useful tool for multiscale design.

Frictional properties of 2D materials are closely watched due to their weak van der Waals (vdW) interlayer interaction, making them excellent candidates for nanoscale solid lubricant. The superlubricity of graphene appears when graphene rotates at certain angles. L. Qiao et al. proposed a DFT calculation combined with ML strategy to investigate the sliding potential energy corrugation between geometrical corrugated graphene sheets. For instance, with the aid of geometrical descriptors and a few DFT calculations of sliding along [110] direction, the trained linear regression models can accurately predict the potential energy evolution along the [100] direction. The supposed strategy could become an effective method to investigate the frictional characteristics of complex interfaces.
3.1.4. Thermal Properties

The thermal conductivity of graphene can reach 5000 W m\(^{-1}\) K\(^{-1}\) with high electron mobility of \(2.5 \times 10^5\) cm\(^2\) V\(^{-1}\) s at room temperature, making graphene and its composites promising thermal interface materials (TIMs) in electronic devices. However, the prediction of lattice thermal conductivity using classical interatomic potentials is still challenging, often observing a variation of one order magnitude compared with experimentally measured values. A. V. Shapeev et al. used MLIPs trained over AIMD trajectories to accurately predict the lattice thermal conductivity of graphene (3600 W m\(^{-1}\) K\(^{-1}\)) which is within the experimentally measured values (1500–5300 W m\(^{-1}\) K\(^{-1}\)). The lattice thermal conductivity of graphene/borophene heterojunctions was also studied and their thermal conductance at the continuum level was further explored by the FEM.\(^{15}\) For the application of graphene-based composites in TIMs, interfacial thermal resistance is an important factor to study heat dissipation at material interfaces. X. Zeng et al. studied the interfacial thermal resistance between graphene and hexagonal boron nitride (hBN) using ML models. The training samples were collected via HTC of MD simulations, considering different combinations of system temperatures, interfacial coupling strengths, and in-plane tensile strains in different directions. As a result, the trained DNN model can accurately predict the interfacial thermal resistance between graphene and hBN with only the knowledge of system temperature, coupling strength, and tensile strain.\(^{16}\)

In practical applications such as temperature sensors, flexible heaters, and wearable e-textiles, graphene sheets are usually deposited and stacked on a substrate. Hence, the thermal properties prediction of stacked graphene sheets with random distributions and arbitrary positions is essential for structural design in application. As shown in Figure 8a, B. Xu et al. developed physics-informed pixel value (PIPVs) matrices (fingerprints) to capture the geometric features of stacked graphene sheets (I). The thermal conductivity of stacked graphene sheets was obtained from MD simulations (II). The gray scale image data serving as input data and the thermal conductivity acting as output data were used to train a DNN to establish the structure–thermal property paradigm (III). The trained DNN accurately predicted the thermal conductivity of stacked graphene sheets (V, IV). A comprehensive databank that stores the geometry features of stacked graphene sheets and their corresponding thermal conductivity was constructed (VI), providing an accelerated search tool to guide the design of stacked graphene sheets (Figure 8b). The developed DNN-based ML can accelerate the structural design of complex systems.\(^{22}\)
The thermal expansion coefficient (TEC) of graphene is another vital property in determining the performance of graphene-based devices. Unfortunately, experimental measurements of thermal expansion coefficient without substrate effects are challenging for 2D materials. MLIPs-based MD simulations have been successfully employed for the theoretical prediction of thermal expansion coefficient. C. Sevik et al. proved that the GAP model catches the lattice dynamics of graphene very well. The estimated TEC value is very close to the DFT-calculated result.[14a]

3.1.5. Cytotoxicity

Since graphene has been used in a variety of applications, determining the cytotoxicity of graphene is essential for environmental and healthy analyses. However, the existing graphene toxicity data are complex and heterogeneous. T. Zhang et al. conducted a meta-analysis of 792 publications studying the cytotoxicity of graphene. Among them, 10 attributes related to the properties of graphene, cell properties, and experimental conditions were extracted. RF algorithms to predict graphene cytotoxicity have been developed based on cell viability, inhibitory concentration (IC50), and lactate dehydrogenase (LDH) release as toxicity endpoints. The exposure dose and detection method of cell viability, diameter and surface modification of IC50, and detection method and organ source of LDH release are the most influential attributes to the cytotoxicity of graphene. The finding indicates that the data mining combined with ML protocol is powerful.[23]

3.2. Structure Recognition

3.2.1. Atomic Structure

Defects (e.g., edges, corners, vacancies, dopants, adsorbates, reconstructions) in graphene and GO have enormous importance on the electronic, optical, thermal, and mechanical properties. The type, density, and distribution of defects play decisive
roles in the properties, thus the defect detection with atomic resolution in graphene and GO is essential to the materials design and discovery problems. However, defects detection by high-resolution experimental characterizations is still technically challenging. ML methods have been used in the atomic-level analysis of graphene and GO, such as defect detection of graphene, bandgap regulation of doped graphene, structural analysis of GO, and the reason explore of fractured C–C bonds in GO.[4b,24] As defect locations are associated with the thermal vibration features, G. X. Gu et al. trained a kRR model by tens of thousands of thermal vibration topographies calculated by MD simulations, discovering the hidden correlation between defect locations and thermal vibration features. As shown in Figure 9a,b, two similar prediction strategies based on different basic units (atom indices, domain discretization) are developed. The atom-based method is used to detect a single-atom vacancy, while the domain-based method can detect an unknown number of multiple vacancies up to atomic precision. In the case of the atom-based method, the vibrational energy distribution throughout the graphene sheet is first computed (step 1); the 2D energy distribution is then compressed to a 1D energy vector with a length of the total number of atom positions, a total of \( \approx 17,000 \) energy vectors are obtained and assembled into a design matrix as the ML training data (step 2); finally, the trained kRR model is used to predict the label vectors which are related to the vacancy locations from unseen energy vectors, and the label vectors are converted to 2D images to receive a better intuition (step 3). The methods achieve a test accuracy of 90\%, indicating a promising extrapolation into other graphene configurations.[4b]

The performance of GO is strongly associated with the oxidation state of the surface, including the number, type, and distribution of functional groups. The Lerf–Klinowski model of GO has been depicted in most publications. However, the model reflects only the chemical connectivity and not the spatial

![Image](https://www.advancedsciencenews.com/)

**Figure 9.** ML-based methods for detecting the structure of graphene and graphene oxide (GO). a) The atom-based method for detecting graphene defects (steps 1~3). b) Domain discretization of a graphene sheet, and the 2D presentation of the label vectors predicted by the domain-based method. Reproduced with permission.[4b] Copyright 2020, Springer Nature. c,d) Representative structures of GO nano flakes: c) the closest matching archetypal GO nano flakes, d) the closest matching prototypical GO nano flakes. Reproduced with permission.[24d] Copyright 2019, IOP Publishing.
distribution of the functional groups, which is inconsistent with the structures obtained by computational studies and experimental observations. Using a large and diverse dataset of 20,936 GO nanoflakes fully relaxed at the electronic structure level, characterized by a total of 830 thermodynamic, structural, chemical, and statistical features, A. S. Barnard et al. determined the truly representative structures of GO using the clustering technique. As shown in Figure 9c,d, only 25 archetypal structures are proposed to capture all the complexity and diversity of the entire dataset, and 3 prototypes are identified to represent the averages of the three classes in the dataset. These 28 structures can be downloaded together as a small dataset with a fraction of the computational costs in future work, or used individually to match experimental observations.[24]

3.2.2. Dimensions and Shapes

Optical micrographs and Raman spectra are widely used to identify graphene domains with a different number of layers in exfoliated graphene flakes and continuous epitaxial graphene films. In addition, the combination of atomic force microscopy (AFM) topography and friction force microscopy (FFM) is also applied for the identification of graphene domains. ML methods (e.g., clustering) allow for building fast and reproducible correlations between topographic data and optical micrographs (or Raman spectra, friction data). The intelligent identification method is beneficial for the identifications of thickness, the existence of impurities, and even stacking order, promoting the development of an efficient and large-area characterization technique of 2D nanostructures.[25] Morphology identification of graphene flakes always relies on the interference color of the SiO$_2$/Si substrate. However, this empirical judgment is sensitive to the thickness of the SiO$_2$ substrate. ML methods can eliminate the identification process, ambiguity in the layer thickness determination, and the parameter tuning process. Tomoki Machida et al. developed a data-driven clustering analysis method to automatically identify the position, shape, and thickness of graphene flakes from the optical microscope images, where clusters owing to the substrate and monolayer, bilayer, and trilayer graphene flakes can be correctly classified into discrete clusters.[25b] W. Zhao et al. applied the SVM algorithm to identify the thickness, impurities, and stacking order of graphene and graphene-based heterojunctions, generating an ML optical identification (MOI) method to map the morphology information to the characteristic color information of the optical photograph.[25c]

Currently, the most widely used synthetic method of graphene and GO sheets is the liquid-phase exfoliation (LPE) method. Since the product quality varies with preparation conditions, the characterization of the liquid-phase exfoliated sheets, nanoplatelets, and flakes of graphitic materials is necessary. The existing imaging tools for visualization of the exfoliated graphene require extensive preprocessing such as centrifugation, membrane dialysis, and flat drying, not available for visualization of graphene dispersed in a liquid. M. Majumder et al. proposed a technique for detecting, classifying, and quantifying the exfoliated materials, utilizing coupled pixel-by-pixel information from bright-field and birefringence images as the training dataset of an unsupervised ML algorithm (clustering). The ML technique can be used to identify three unique species, including flakes (unexfoliated), nanoplatelets (partially exfoliated), and 2D sheets (well-exfoliated) in dispersions of graphitic materials. Undertaking the pixel-by-pixel analysis, the size, thickness, and concentration of these exfoliated species can be further measured. The approach can quantitatively assess one sample of graphene/GO dispersion on short notice, serving as an efficient in situ classification and quantification tool in industrial-scale production.[26]

Twisted bilayer graphene (tBLG) has aroused wide attention in the past years, sparked in part by the finding of superconductive states at small twist angles. Raman spectroscopy provides a method to measure the twist angle. However, changes in the Raman spectra induced by the stacking order can be very subtle, and the Raman signal can also be altered by the states (e.g., defects, strain, doping) of graphene, making the identification task difficult. ML approaches are well adapted to find a quantitative method to characterize tBLG using Raman spectroscopy, that is the correlation between Raman spectra and the twist angle of tBLG can be built using supervised ML models.[27] H. Ago et al. proposed an RF classifier (RFC) to automatically classify the Raman spectrum of tBLG into a selected range of twist angles. The training dataset is generated from ≈6000 individual Raman spectra collected from different graphene samples, and the ML method extracts features of the Raman spectrum of tBLG to train an ML model (Figure 10a). The trained model is then ready to predict the twist angle of tBLG from its Raman spectrum (Figure 10b–d). The pair of features (I$_{2DC}$ and FWHM$_{2D}$) with the largest values of mutual information can be used to visualize the decision boundary of the model (Figure 10e). The model prediction of a tBLG area coincides with the manual classification with an accuracy of ≈99% (Figure 10f,g). The method provides predictions for whole Raman mappings consisting of hundreds of spectra in a matter of seconds even on average desktop computers, indicating a useful and simple analysis tool with practical applications in the nascent field of twistronics.[27a]

3.3. Inverse Design

Compared with solely predicting the properties of known materials, designing new materials with expected properties is more important. The former is regarded as a forward model problem where the properties of materials are determined by their structures, while the latter is an inverse design problem that generates the structures with desired properties. The QSPR used for properties prediction is first built by ML models, plenty of candidates (unexplored) are then screened through the QSPR to perform inverse design.[28] Once the topological or geometrical features are encoded by a set of structural fingerprints or descriptors, the material property is mapped into structural features. The resulting QSPR is efficient to discriminate potential nanomaterial candidates from virtual graphene libraries or highlight the most influential structural features for structural design. Therefore, the ML technique can efficiently perform the reverse design.[12] A classic example is the structure and composition design of composites which is extremely challenging due to numerous combinations of materials and geometrical configurations. G. X. Gu et al. obtained a tougher and stronger
graphene/graphane (hydrogenated graphene) composite using an ML model that learns the QSPR from a small amount of training data. All the optimal designs of the composites with different volume fractions are shown in Figure 11a, and their toughness indicates the optimal volume fraction of the toughest composite is 40.6% (Figure 11b). The optimal graphene/graphane composite can store more elastic energy before failure (Figure 11c).

The ML technique can also extract the most influential factors controlling the properties to guide the design of materials. A. Fazzio et al. combined DFT calculations with ML models to evaluate the chemical and structural relevant parameters governing the binding strength between GO and nanocellulose. The oxygenated group density of GO is the primary attribute ruling the binding energy scale. As a result, the refined control over the binding energy based on the variation of the oxygen density in GO is an effective way.

The microstructure of graphene can modulate the electronic, thermal, and mechanical properties, such as opening a finite bandgap, reducing the thermal conductivity, and optimizing the mechanical properties. Many microstructural parameters, such as the edge topology and overall shape of pristine graphene and the density and spatial distribution of holes in porous graphene, play an important role in the design of graphene, indicating a large design space. Making kirigami-inspired cuts into graphene sheets is effective to design stretchable graphene sheets with metamorphic properties, and rational cutting patterns are essential to obtain the target properties of graphene sheets. H. S. Park et al. utilized a CNN model to construct the QSPR between cutting patterns and mechanical properties, searching for optimal kirigami patterns with extreme stretchability in a large design space of \( \approx 4 \times 10^6 \) candidate designs (Figure 11d,e). The kirigami structures found here using ML are also applicable for designing larger macroscale kirigami structures. In another case, H. S. Park et al. introduced holes into graphene sheets to tune the thermal conductivity, designing the optimal porous graphene sheets with the lowest thermal conductivity.

3.4. Task Recognition

3.4.1. Graphene-Based Sensors

Graphene is widely used as a sensing material of sensors (especially wearable sensors), because of the large surface area, unique optical properties, excellent electrical, thermal, and mechanical (e.g., strength, flexibility) properties. For instance, the large specific surface area is responsible for the high sensitivity of electrochemical sensors. The high mechanical flexibility makes sensors highly wearable. The high strength and electrical conductivity enable faster signal response, higher maximum sensing range, and better response reproducibility of strain sensors. The changes in electrical, mechanical, and photophysical
properties of graphene and its composites can be used as the transduction mechanism of sensors. Various ML techniques have recently been integrated with graphene-based sensors for data analysis, classification, and diagnosis. The sensor combined with ML approach enables faster and more diverse detection or recognition of chemicals and motions. Particularly, the cutting-edge approach will bring a revolutionary breakthrough in the rapid development of advanced wearable electronics in the field of human health monitoring.

3.4.2. Chemical Recognition

When the human body touches an object, the temperature, wetness, and types of materials with different thermophysical properties can be perceived. Graphene strain sensors coupled with ML system represents a great artificial tactile system to identify material species and micro-sculpture patterns, showing a complete superiority to human fingers. While mechanical tactile perception has been widely demonstrated in artificial sensors, the indirect sensing of thermosensation is based on learning and experience. Emulation of the human somatosensory system using artificial sensors and ML algorithms enables enhanced environment and object recognition in the application of robots, wearables, and haptic interfaces. H. Ko et al. introduced an artificial somatosensory system based on RGO composites to emulate the somatosensory system of human skin. The artificial somatosensory system coupled with an ML algorithm can differentiate types of contact materials (e.g., organic solvents) with different thermophysical properties (e.g., thermal conductivity, heat capacity, vapor pressure, and heat of vaporization). In the case study about the objective evaluation of the thermal attributes (coolness and wetness) of skincare products (Figure 12a), graphene composites-based sensors can monitor the temporal electrical resistance changes caused by the applied skincare products (i). The data of temporal variations in the resistance are treated by regression analysis to generate variables (input) and the corresponding scores (output), using as the training datasets of ML (ii, iii). The trained extreme gradient boosting (XGBoost) model is then used for evaluating scores about the coolness and wetness of skincare products (iv). In another
case, an array of semi-selective chemical sensors that respond to many chemicals simultaneously is essential to the detection and discrimination of a broad range of chemicals. The data collected by the array-based sensors is then analyzed using ML algorithms for subsequent detection and discrimination. J. R. Uzarski et al. used 12 different polymer–graphene nanoplatelets composite coatings to create an array of semi-selective sensors to detect 13 chemicals. The generated dataset is processed and dimensionally reduced using principal component analysis (PCA). Four algorithms, including kNN, SVM, RF, and LDA, are used to classify chemicals with the classification accuracies of 95%–99%.\[3\]

Gas sensing is essential for many applications, such as environmental monitoring, drug screening, medical diagnosis, food storage, and alcohol testing. For example, detecting volatile organic compounds (VOCs) in human breath is critical for the early diagnosis of diseases, air quality monitoring is extremely important to health care. The interactions of chemical vapors with sensors generally take place on the surface of the sensing layer, where chemicals are adsorbed at the surface or interface. Artificial olfactory systems, namely electronic noses or e-noses, benefit from their gas selectivity. The e-nose system has artificial components like the olfactory organ, comprising an array of gas sensors, output vectors, and a pattern recognition algorithm. The output vectors are projected to feature space for gas classification.\[3b,34\] Learning from the concept of e-nose, L. Lin et al. reported a novel gas-sensing scheme using a single graphene FET (GFET) and an ML model to realize gas identification. The gas-sensing conductivity profiles of a GFET are recorded and decoupled into four distinctive physical properties (Figure 12b), namely projected onto a feature space as 4D output vectors. The detected gases and corresponding 4D output vectors are correlated using the MLP classifier which can then be used to classify water, methanol, and ethanol vapors with high accuracy.\[34\] Similarly, many studies use the ML–PCA method to realize the separation of different types or concentrations of chemical gases in space coordinates based on principal components. These gas sensors can be used as small, low-power, and robust e-noses.\[3b,35\]

3.4.3. Motion Recognition

A human–machine interface (HMI) is a bidirectional electronic system that connects humans and machines to control hardware and collect feedback information. The HMI has been widely used in many applications such as the humanoid robot, prosthetic hand, display interface, electronic wheelchair. Wearable HMI systems are getting increased attention with the development of wearable electronic devices. The graphene-based sensors combined with ML systems have been widely used as wearable HMI systems for applications in motion recognition.\[3c,36\] As shown in
Figure 13. S. O. Kim et al. combined a large-area pressure sensor array with an ML algorithm to construct the smart seat cushion-based posture monitoring system. Graphene-MXene hybrids are utilized as sensing materials for piezoresistive pressure sensors with low hysteresis and a wide sensing range. Large-area pressure sensor arrays are integrated into seat cushions for healthcare monitoring. The variation in resistance is monitored in real-time at each pixel for different seating postures. RF and ANN models are trained using the collected data, differentiating 6 sitting postures with high accuracy. Similarly, W. H. Yeo et al. used biocompatible solderable graphene to fabricate an all-printed wearable and wireless device which incorporates ML algorithms to implement multi-class and versatile HMI scenarios. ML algorithms (CNN, kNN) trained using the measured electromyogram data successfully detect 7 classes of finger motions with an accuracy of ≈99%. Speech-based HMI is on the rise as well because it allows users to interact with computers using only their voice, which is especially helpful for people who have difficulties in typing on keyboards and mice. In addition, silent speech recognition which can recognize intended speech without audio information is important for people who have hearing and speech impairments. C. Malia et al. developed an approach to classify recorded resistance signals into predicted words. A graphene strain gauge sensor is first fabricated and worn on the throat to detect signals of small muscle movements and vocal vibrations as the training dataset of ML. ML techniques (RF, kNN, ANN) are then used to analyze the relationship between the detection signals and intended speech, and thus make correct predictions of speech with previous unseen signals.

3.4.4. 3D Imaging

Recently, optical imaging which extracts 3D information from what is normally a 2D image capture is developed rapidly. The 3D imaging is particularly important for applications in autonomous vehicles, face recognition, unmanned aero vehicle navigation, and biological video-rate 3D microscopy. T. B. Norris et al. demonstrated 3D tracking of point-like objects with multi-layer feedforward neural network and the extension to track positions of multi-point objects. A transparent focal stack imaging system is built using graphene photodetector arrays (Figure 14a,b). In the typical imaging process, the camera lens...
projects an arbitrary object onto a set of transparent imaging arrays stacked at different focal planes. Each of the images in the stack records the light distribution using graphene photodetector arrays at a specific depth. The ANN model is then used to process the 3D focal stack data and estimate the 3D position and configuration of the object. The 3D tracking of a ladybug is taken as an example (Figure 14c,d). The traditional complementary metal–oxide–semiconductor (CMOS) sensors are first used to record 15,488 high-resolution focal stack images, and two CNN algorithms are then trained by these images to precisely estimate the ladybug’s position and orientation.[36]

4. Graphene-Based Artificial Synapses

Emulating biological synapses using electronic devices is an important research frontier in modern technology, which is one of the key components in hardware building brain-like computers and AI systems. In the past decades, silicon-based transistors and CMOS artificial circuits have been used to emulate synaptic functions because of their good stability and controllability. However, the traditional semiconductor materials are approaching their physical limits in Moore’s law. The 2D materials have shown a unique immunity to short-channel effects and have been applied in neuromorphic devices. What’s more, 2D materials can be used to fabricate flexible ASs, which can be combined with sensory modules to realize the similar functions of biological sensory systems and even motor nerves. Graphene has the advantage of high mobility, high transparency, excellent thermal stability and conductivity, high mechanical properties, and large theoretical specific surface area. In addition, graphene has a unique characteristic of zero-bandgap allowing a practical gate voltage to tune the transition between electron- and hole-dominated conduction in graphene. Owing to these merits, many synaptic graphene transistors have been proposed to emulate biological synaptic plasticity such as potentiative and depressive plasticity, and modulatable plasticity.[37]

In contrast, two-terminal memristors have a simple structure and show low energy consumption. Memristors have shown excellent synaptic behaviors by gradually changing their resistance states under the electrical input stimuli. The memristive materials of synaptic memristors are usually transition metal oxides. However, the corresponding devices exhibit a high programming current and a high RESET current in the range of μA and mA, which will increase the energy consumption of the device. Recently, 2D materials have been used in memristive synapses owing to their excellent physical properties, high electrical tunability, low-power-switching capability, and heterointegration compatibility. Graphene has been used in...
memristors due to its excellent intrinsic characteristics (e.g., flexibility, low cost, adaptability, and environmentally friendly), serving as memristive materials or back electrodes.[39]

4.1. Principles and Methods

For a synaptic graphene FET, the graphene channel and gate dielectric layer perform the synaptic transmission between two neurons. The gate electrode functions as the presynaptic membrane receiving electric signals, while the drain electrode works as the postsynaptic membrane generating action potentials. The conduction state of graphene can be modulated by the gate voltage due to its zero-bandgap feature. When a certain bottom gate voltage \( V_{bg} \) or the top gate signal (e.g., top gate voltage, \( V_{tg} \)) is applied, the graphene channel becomes hole- or electron-doped, resulting in the nonvolatile displacement of the Dirac point and an obvious hysteresis window in the conductivity (Figure 15a). As shown in Figure 15b, once the voltage is applied to the gate electrode, the induced carriers generate a sudden drain current \( (I_d) \) change (e.g., points 1 \( \rightarrow \) 2 or 3 \( \rightarrow \) 4), the subsequent current change is due to the trap or release of carriers in graphene (e.g., points 2 \( \rightarrow \) 3 or 4 \( \rightarrow \) 1). The current change between points 1 and 3 reflects the change of PSC, namely the synaptic plasticity. The direction of applied gate voltage determines the polarization of APSC which shows potentiative or depressive plasticity. When the channel material is a heterojunction, an obvious hysteresis window of the Dirac point and carrier concentration variations in the synaptic memristor through various mechanisms such as defect migration, vacancy migration, filament formation, and charge trapping represent the changes in synaptic weight. As shown in Figure 15c, memristors present a unique voltage–current relationship determined by a closed hysteresis loop pinched in the origin. By increasing the bias, the device transforms from an HRS (segments 1 and 2) to an LRS (segment 3), then maintains the LRS (segments 3 and 4) and finally returns to an HRS (segment 5). The positive and negative currents of the device change gradually with the change in voltage, serving as an important basis for bidirectional resistance control of a bio-synapse emulator. The resistance state of the insulating material varies between LRS and HRS, resulting in two operations: SET (HRS to LRS) and RESET (LRS to HRS).[7a,7b] In addition, memristors usually have multiple

Figure 15. Measurement principles and methods of artificial synapses (ASs). a) Electric hysteresis loop of field-effect transistor (FET) by sweeping \( V_{tg} \). Reproduced with permission.[5a] Copyright 2019, Springer Nature. b) Hysteric behaviors of FET labeled with the sweeping direction. Reproduced with permission.[5c] Copyright 2021, IOP Publishing. c) Current–voltage curve of memristor (inset: the corresponding logarithmic representation). Reproduced with permission.[7b] Copyright 2020, Royal Society of Chemistry. d) Manipulation of the memristor resistance with positive and negative voltage pulses. Potentiation (red dot) is achieved with a positive pulse, while depression (black dot) is achieved with a negative pulse. e) LTP and STP. f) paired-pulse facilitation (PPF) and paired-pulse depression (PPD). g) Typical STDP according to the relative timing of presynaptic and postsynaptic pulse. h) SRDP for LTP and LTD at different frequencies. Reproduced with permission.[41] Copyright 2021, John Wiley and Sons.
conductance states similar to the weight adjustment in biological synapses. The resistance or conductance changes gradually with the increase of pulse number (Figure 15d).

Through the regulation of the number, frequency, and polarization of the discharge pulse, the synaptic transistors and memristors can emulate various synaptic behaviors, such as STD, STP, LTD, LTP, SRDP, STDP, paired-pulse facilitation (PPF) and paired-pulse depression (PPD). 1) The synaptic weight of ASSs changes with periodically current pulses, then returning to the initial value after a short or long period of time, corresponding to STP/STD and LTP/LTD, respectively (Figure 15e); 2) STP can be further divided into PPF and PPD (Figure 15f). When a series of spike signals are transmitted close enough to affect each other, the short-term synaptic weight of PPF is enhanced, while PPD does the opposite. The paired pulse ratio (PPR) is the response intensity ratio of the second and first stimuli ($A_2/A_1$); and 3) The synaptic weight is dependent on the time interval between the presynaptic and postsynaptic spikes and related to the frequency of the individual spikes, corresponding to STDP and SRDP, respectively (Figure 15g,h). [5b,5e,7b,41]

### 4.2. Transistors

Transistor-based three-/multi-terminal devices are stable and relatively controllable, and can be constructed by various materials. The transistor-based ASSs perform concurrent learning, namely updating synaptic weight without interrupting the signal transmission process. They can also realize the synergistic control of one device. Through rational materials design, these devices convert various external stimuli into electric signals.

#### 4.2.1. External Powered Systems

The gate dielectric layers of traditional transistors are usually high-k solid-state materials. [42] Recently, many dielectric materials, such as ionic gels, [5a,43] plasma, [44] ferroelectric materials, [5b] and other solid electrolytes (e.g., SiN$_x$H$_y$), [45] are used as gate dielectric layers in synaptic graphene transistors. Especially, ion-gel has been widely used in graphene transistors due to its faster ionic mobility and diffusivity, and easy preparation. The synaptic weight of the synaptic graphene FET is controlled by applying a gate voltage. [5a] X. S. Miao et al. designed a ferroelectric-gate synaptic transistor using graphene as the channel and polyvinylidene fluoride (PVDF) ferroelectric polymer as the gate dielectric layer (Figure 16a). The conductance of graphene can be tuned through the polarization of the ferroelectric layer (Figure 16b). A large negative/positive gate voltage over the coercive voltage result in upward/downward polarization of the PVDF dielectric layer, leading to a hole/electron dominated graphene channel due to different positions of Fermi levels within the energy bands of graphene. Further, the addition of a small positive gate voltage will decrease/increase the upward/downward PVDF polarization, leading to the upward shift of the Fermi levels in both p- and n-type graphene channels. In this way, the analog weight update of the corresponding AS can be potentiative or depressive when imposing similar programming gate voltages (Figure 16c,d). In addition, the nonvolatile nature of ferroelectric polarization causes persistent and memorable modulation of the ferroelectric synapse, showing synaptic plasticity of LTD and LTP. [5b]

The design of channel materials is also crucial to improve the efficiency, tunability, stability, and symmetry of the synaptic plasticity of synaptic transistors. The tBLG has been used for replacing the graphene channel to make an artificial dynamic synapse, realizing synaptic plasticity, and dynamics simultaneously. [54] In addition, the combination of graphene with other 2D materials (e.g., MoS$_2$/graphene, WSe$_2$/graphene, fluorographene/graphene) [5c,5e,39b] or oxide semiconductors (e.g., InGaZnO/graphene) [46] to form compound channel materials is not only beneficial for modulating synaptic plasticity through heterojunction or polarization but also effective to realize the synaptic dynamics. In short, this method provides a multidimensional modulation strategy to regulate the synaptic plasticity and dynamics of synaptic transistors under stimuli. The achieving of a stable and long-term weight-modulated synaptic graphene transistor is still challenging due to the nature of the spontaneous relaxation of interface charge storage. The stable and tunable dipolar polarization in fluorographene can modulate the analog conductance of the synapse, thus demonstrating long-term plasticity. C. S. Lai et al. proposed a fluorographene/graphene (FGr/Gr) synaptic transistor. The Al gate and Ni electrode serve as the pre-synapse and post-synapse membranes, respectively, and FGr/Gr or Gr serves as the synapse. After 50 successive positive voltage pulses, the FGr/Gr synaptic transistor (FGr-ST) exhibits higher variation efficiency and more stable

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**Figure 16.** Synaptic graphene transistor with a ferroelectric dielectric layer. a) Schematic of the device (left) and the polyvinylidene fluoride (PVDF) ferroelectric polymer (right). b) Schematic view of tuning the FET to be potentiative synapse (up) or depressive synapse (down). c,d) Analog weight update of synapses: c) depressive, d) potentiative. Reproduced with permission. [Copyright 2019, Springer Nature.]
memory (lasting 200 s without relaxation) than the Gr synaptic transistor (Gr-ST). The tunability of Gr-ST is induced by charge trapping and spontaneous detrapping at the dielectric and channel interface. While the tunability for FGr is mainly due to the modulation of the C–F dipolar orientation and its local electric field, thus resulting in a stable and long-term weight modulation in the FGr-ST device.[5c]

With the rapid process of imaging technology, the dynamic behavior in biological synapses has aroused widespread interest. The realization of the conversion between inhibitory and excitatory synapses in a dynamic synaptic device can mimic the behaviors of living organisms in higher dimensions. The modulation of synaptic plasticity not only raises the abundance of synaptic plasticity but also holds possibilities for developing neuromorphic systems with autonomous learning and self-healing abilities. To realize the tunable plasticity of AS, T. L. Ren et al. paired tBLG and Al2O3 as a function layer to fabricate a graphene transistor with a bottom gate (Figure 17a). The independent layers of tBLG can separate the effects of the two gates (top and bottom), leading to an easier shift of the hysteresis curve and the intersection transfer between two branches by changing the $V_{bg}$ (Figure 17b). A relatively intact process of synapse development is shown in Figure 17c, $V_{bg}$ is the factor that controls the conversion between inhibitory and excitatory synapses.[5d] In another case, the asymmetric optical response of heterojunctions allows the AS device to show both electrically and optically modulated synaptic plasticity. T. Palacios et al. designed an artificial synaptic transistor based on a WSe$_2$/graphene heterojunction. The analogy between a synaptic WSe$_2$/graphene transistor and a biological synapse is shown in Figure 17d. The excitatory–inhibitory plasticity conversions can be modulated by a variety of electrical signals, including drain voltage ($V_{ds}$), the polarity of the gate voltage ($V_{gs}$) spikes, and $V_{gs}$. The $V_{gs}$ spike polarity or $V_{gs}$ value-dependent synaptic plasticity is attributed to the changes in energy band caused by the $V_{gs}$ regulation (Figure 17e). While the change of $V_{ds}$ can lead to the shift of the hysteresis curve and the intersection transfer induced by the voltage-dependent rectification characteristics of the heterojunction. In addition, optical stimuli can be used as another physical domain of modulation to realize the conversion between inhibitory and excitatory synapses. The optical modulated synaptic plasticity can be explained by the photoinduced shift of the hysteresis curve and the intersection transfer.[5e]

4.2.2. Self-Powered Systems

Self-powered transistors can be realized by using piezotronic or triboelectric materials as gate dielectric layers, which generate electricity by the piezoelectric or triboelectric effect to provide gate voltage to the graphene channel, respectively. For instance, piezotronic-gate transistors utilize piezopotential as the gate voltage to modulate the charge carriers’ transport at the electrical

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**Figure 17.** Synaptic transistors based on different graphene-based channel materials. a) Schematic of a tBLG dynamic synapse. b) $I_d$ versus $V_g$ under the $V_{bg}$ of $+40$ V (left) and $-40$ V (right). c) Development of the dynamic synapse, steps 2 ~ 4 and 6 ~ 8 represent the formation, maturation, and elimination of excitatory synapse and inhibitory synapse, respectively. Reproduced with permission.[5d] Copyright 2015, American Chemical Society. d) An analogy between a WSe$_2$/graphene heterojunction-based synaptic transistor and biological synapse. e) Band alignment of graphene/WSe$_2$ under $V_{gs}$ of 25 V (left) and $-25$ V (right). Reproduced with permission.[5e] Copyright 2021, IOP Publishing.
junctions. However, a more general piezotronic AS device is defined as an electronic transistor modulated (or powered) by a piezoelectric nanogenerator (PENG). The coupling between piezoelectric polarization and semiconducting transport properties offers an active and direct trigger of external pressure with electrical output signals. The synapse composed of a transistor and a nanogenerator is a typically self-powered system. Nanogenerators can convert mechanical signals into electrical signals. The electrostatic potential generated by nanogenerators can regulate channel conductance of traditional transistors and ionic liquid-gated or gas-ions-gated transistors. For the latter, ionic liquid or plasma replaces the high-k solid-state materials as gate dielectric layers. By regulating the number, frequency, and polarization of the force pulse, various synaptic behaviors can be achieved, such as STD, STP, LTD, LTP, SRDP, STDP, and PPF. The neural functions of learning and temporal decoding can also be realized.  

Q. Sun et al. demonstrated a piezotronic graphene AS device by integrating a PENG with an ion-gel-gated graphene transistor (Figure 18a). The piezopotential of the PENG results in the formation of electric double layers at the interface of the ion gel/electrode and ion gel/graphene (Figure 18b). In the stretch mode, the Fermi level of graphene is shifted downward and the holes in the channel dominate the charge transport, leading to an increased output current. In contrast, the compression strain results in a slight decrease in output current. This design can also be applied to mechno-photonic AS devices composed of optoelectronic transistors and nanogenerators. Once employing the triboelectric potential to control the charge transfer in the transistor, the optoelectronic synaptic behaviors can be modulated. Z. L. Wang et al. presented a bioinspired mechno-photonic AS with synergistic mechanical and optical plasticity (Figure 18c). The AS is composed of a graphene/MoS2 transistor and an integrated triboelectric nanogenerator (TENG). The triboelectric potential provided by TENG displacement effectively drives the transistor and modulates the charge transfer/exchange in the graphene/MoS2 heterostructure (Figure 18d), thus leading to triboelectric potential-modulated electrical behaviors (Figure 18e). The electrical characteristics under the separation state ($D^+$) and a single light pulse are similar to the LTD behavior in the biological synapse.

More structures and performance parameters of synaptic graphene transistors are shown in Table 1.
Table 1. Synaptic graphene transistors.

| Device                              | Device size | Stimuli          | Mechanism                     | Synaptic functions | Spike attenuation | Endurance | On/off ratio | Energy (per spike) | References |
|-------------------------------------|-------------|------------------|-------------------------------|--------------------|-------------------|-----------|--------------|-------------------|------------|
| AIOₓ-gated graphene transistor      | N/A         | Electric field   | Surface charge trapping       | STD/STP/LTD/LTP/PPF | N/A               | 54.86 s   | N/A          | N/A               | [5c]       |
| Silver gel/polarized aptamer-gated  | Lᵢₑᵢ = 800 μm | Electric field   | Electric polarization         | STD/STP/LTD/LTP/PPF | 0.5~10 s          | >5 min    | N/A          | 20 pJ             | [5a]       |
| graphene transistor                 | Wᵢₑᵢ = 800 μm | Electric field   | Electric polarization         | STD/STP/LTD/LTP/PPF | 0.5~2 s           | N/A       | N/A          | N/A               | [43a]      |
| Ion-gel-gated graphene transistor   | Lᵢₑᵢ = 70 μm | Electric field   | Electric polarization         | STD/STP/LTD/LTP/PPF | N/A               | N/A       | 2.2          | 50 nJ             | [5b]       |
| PVDF-gated graphene transistor      | Lᵢₑᵢ = 85 μm | Electric field   | Electric polarization         | STD/STP/LTD/LTP/PPF | N/A               | >200 s    | N/A          | N/A               | [5c]       |
| Ion-gel-gated RGO/InGaZnO transistor | N/A         | Electric field   | Electric polarization         | STD/STP/LTD/LTP/PPF | N/A               | N/A       | N/A          | N/A               | [46]       |
| AIOₓ-gated fluorographene/graphene  | N/A         | Electric field   | Electric polarization         | STD/STP/LTD/LTP/PPF | N/A               | N/A       | N/A          | N/A               | [5c]       |
| transistor                           |             |                  |                               |                    |                   |           |              |                   |            |
| Perovskite quantum/ h-BN/graphene   | Lᵢₑᵢ = 200 μm | Light            | Photoelectric effect          | STD/STP/LTD/LTP/PPF | >15 s             | >60 s     | N/A          | N/A               | [47]       |
| transistor                           | Wᵢₑᵢ = 500 μm |                  |                               |                    |                   |           |              |                   |            |
| PENG/ion gel-gated graphene transistor | Lᵢₑᵢ = 60 mm | Pressure          | Piezotronic effect            | STD/STP/LTD/LTP/PPF | 1.24~3.24 s       | N/A       | N/A          | N/A               | [39a]      |
| transistor                           | Dᵢₑᵢ = 10 μm |                  |                               |                    |                   |           |              |                   |            |
| TENG/gas-ions-gated graphene        | Lᵢₑᵢ = 40 μm | Pressure          | Triboelectric effect          | STD/STP/LTD/LTP/PPF | 11 s              | 32 h      | N/A          | N/A               | [44]       |
| transistor                           | Wᵢₑᵢ = 10 μm |                  |                               |                    |                   |           |              |                   |            |
| TENG/graphene/MoS₂ transistor        | Lᵢₑᵢ = 20 μm | Light            | Triboelectric effect          | LTD/LTP            | N/A               | >1 h      | 590~857      | N/A               | [39b]      |
| TENG/ion gel-gated graphene transistor | Lᵢₑᵢ = 300 μm | Magnetic field   | Triboelectric effect          | LTD/LTP            | N/A               | >1 h      | 590~857      | N/A               | [39b]      |

4.3. Memristors

A memristor is a resistor with memory, emerging as a resistive random-access memory (RRAM) type technology beyond CMOS-based platforms. Memristors-based two-terminal devices have the advantage of scalability, low energy consumption, and ultrafast data-processing speed.

4.3.1. Graphene Memristive Layer

Graphene-based memristors can be classified into lateral-, vertical-, and heterojunction-structures. These devices operate with different mechanisms depending on their structures and materials.[38] In lateral-structure devices, charges flow along the in-plane direction. Defect migration or phase transition results in resistance change.[46,47] N. Rodriguez et al. proposed the fabrication of laser-lithographed GO memristors. The laser triggers a photothermal reduction process that removes oxygen-containing functional groups and partially recovers the sp² hybridized C-C bond of GO.

Both ends of the prepared GO memristive layer are led by Ag-based conductive paint to fabricate a lateral-structure memristor. The physical mechanism of the RS process is attributed to the drift of oxygen ions and oxygen-containing groups inducing local changes in the level of reduction of the GO (Figure 19a). Low-resistance conductive paths are formed in the bulk of the material when sp³ domains are turned into sp² domains.[64]

In vertical-structure devices, charges flow along the out-of-plane direction. General memristive switching mechanisms contain phase transition, vacancy migration, filament formation, and transport conversion from Schottky emission (SE) to direct tunneling (DT).[6b,49] C. Choi et al. prepared a biocompatible/organic synaptic memristor based on an N-doped graphene oxide quantum dots (N-GOQDs) insulating layer, that is, the Ag/N-GOQDs/Pt–Ti memristor (Figure 19b). The Ag⁺ migration dynamics mechanism of the memristor is in analogy to the Ca²⁺ ion dynamics of biological synapses. The programming/erasing processes of the memristor are realized by the oxidation/reduction chemical reactions of Ag filament inside the
Figure 19. Synaptic memristors with GO memristive layers. a) The working mechanism of a laser-fabricated GO memristor with a lateral structure. Reproduced with permission. Copyright 2019, MDPI. b) The preparation process of an Ag/N-doped graphene oxide quantum dots (N-GOQDs)/Pt memristor with a vertical structure (right). c) The working mechanism of Ag\(^{+}\) cations migration in the N-GOQDs layer of the Ag/N-GOQDs/Pt memristor. Reproduced with permission. Copyright 2019, John Wiley and Sons.

Figure 20. Synaptic memristor with a graphene/MoS\(_2\)/xO\(_x\)/graphene memristive layer. a) Schematic of the device. b) Optical microscope image and measurement setup of the device. c) Microscopic cross-sectional image of the memristive layer. d–f) Schematic diagrams illustrating the resistive switching (RS) mechanism in the device: d) the pristine state, e) ON state, and f) OFF state. Reproduced with permission. Copyright 2018, Springer Nature.
N-GOQDs layer (Figure 19c). Strong or repetitive pulses inputs are required to build thick/strong and stable Ag filament in the N-GOQDs layer and thus implement the transition from STP to LTP.\[^{6b}\]

In heterojunction-structure devices, the memristive layer consists of 2D–2D or 2D–3D heterojunctions. The devices operate based on vacancy migration, filament formation, and charge trapping mechanisms.\[^{6c,50}\] Stacking 2D materials together can combine the properties of each 2D component. The excellent structural stability of 2D material is beneficial for improving the robustness of electronic devices. As shown in Figure 20a–c, F. Miao et al. proposed a high thermal stability memristor based on the graphene/MoS\(_2\)/Mo\(_x\)O\(_x\)/graphene vdW heterostructure. The device shows repeatable bipolar RS performance, possesses a high on/off ratio and switching speed, and remains fully functional at 340 °C. The RS switching mechanism is primarily based on the migration of oxygen ions with minor structure changes of the channel region (Figure 20d–f)\[^{6c}\].

### 4.3.2. Graphene Back Electrodes

Graphene electrodes have a high out-of-plane contact resistance and weak vdW forces, beneficial to reduce the running current, increase the on/off resistance ratio, and lower the energy consumption of a memristor.\[^{7}\] C. S. Lai et al. employed a graphene back electrode to fabricate a graphene-based neuromorphic memristor with programmable metaplasticity (Figure 21a). In response to the voltage stimuli, the resistance of the memristor changes due to the field-induced reconfiguration of oxygen vacancy in the AlO\(_x\) insulating layer. Through controlling compliance current (CC) which can be seen as the historical stimulation, the memristor exhibits programmable

![Graphene Back Electrodes](image)

**Figure 21.** Memristor made of a graphene back electrode. a) Schematic illustration of the memristor (left), and microscopic image of the memristor (right). b) Schematic illustration of the concept of programmable metaplasticity. c) Synaptic plasticity of the memristor with a prior setup of 50 μA (left) and 50 nA (right) compliance current (CC). Reproduced with permission.\[^{7a}\] Copyright 2018, American Chemical Society.
4.3.3. All-Carbon Memristors

As mentioned earlier, many graphene-based synaptic memristors are the combination of graphene with non-carbon materials. The non-carbon materials limit the development of flexible and wearable memristors. All-carbon memristive synapses are promising for future wearable NC systems. The synapses can be transferred onto diverse substrates, showing good flexibility and transferability. They also exhibit high thermal and chemical stability, applicable for different application environments. Unfortunately, GO-based memristors generally show digital-type RS (D-RS) between LRS and HRS via the migration of its intrinsic oxygen functional groups. The realization of analog-type RS (A-RS) with continuous resistance-state variations is essential for complex NC systems. Y. Liu et al. proposed a GO-based all-carbon memristive synapse with A-RS characteristics (Figure 22a). An RGO layer and graphene layer (on a Cu supporting substrate) serve as the top and bottom electrodes, respectively. A composite film of GO and N-doped carbon quantum dots (NCQDs), namely GO-NCQDs, acts as the switching layer. Ultraviolet light irradiation is then introduced to induce the local reduction of GO near the NCQDs. As shown in Figure 22b, multiple weak conductive filaments preferentially grow around the RGO domains under the electrical stimulus, resulting in A-RS. Differently, the D-RS is generally attributed to the formation and rupture of a dominant conductive filament inside the GO. As a result, the RGO/GO-NCQDs/graphene memristor with A-RS characteristics shows much higher linearity in synaptic weight changes (Figure 22c). The all-carbon memristive synapse is beneficial for exploring wearable NC systems in the future.

More structures and performance parameters of synaptic graphene memristors are shown in Table 2.

5. Summary and Outlook

Two strategies, including ML (software) and NC (ASs and artificial neurons) (hardware), have been proposed to imitate the function of a biological brain that efficiently performs the real-time processing of unstructured data. ML has made remarkable progress in properties prediction (electrical, mechanical, thermal, cytotoxicity), structure recognition (atomic structure, microscopic dimensions/shapes), inverse design (composition, microstructure), and task recognition (chemical recognition, motion recognition, 3D imaging) of graphene and its composites. 1) ML-assisted theoretical calculation (DFT, MD) not only enables high-precision properties prediction with a fraction of computational costs but also benefits to studying the material properties under complex factors. Since the actual electronic, mechanical, and thermal properties of graphene affected by preparation and test conditions are much lower than its theoretical properties, combining ML with experimental results for actual properties prediction is important in future studies; 2) The application of ML in structure recognition is effective to interpret differences in properties without sophisticated atomic-level characterization. The combination of ML and material characterization techniques will accelerate the characterization of graphene with high accuracy, especially the large-area characterization; 3) ML is essential for inverse design which determines the optimal design of materials with expected performance or extracts the most influential factors to guide the design of materials, getting rid of traditional trial-and-error research. However, the real inverse design is still under concern due to its screening mechanism, unavailable to derive the best material components only from the ML model trained by the training dataset; and 4) ML has been integrated with graphene-based sensors for data analysis, classification, and diagnosis. Nevertheless, the prediction range and accuracy are limited, only a few scenarios with limited accuracy are considered. In addition, 3D imaging achieved by graphene photodetector arrays is attractive, more research should be undertaken.
Table 2. Synaptic graphene memristors.

| Device          | Mechanism                  | Synaptic functions       | $V_{set}/V_{reset}$ | On/off ratio | $t_{set}/t_{reset}$ | Endurance | SET/RESET energy | References |
|-----------------|----------------------------|--------------------------|---------------------|--------------|---------------------|-----------|------------------|------------|
| Ag/N-GOODs/Pt   | Filament formation Graphene: insulating matrix | STP/LTP/LTD/STDP | 1 V/−0.2 V | 10$^7$ | 200 μs/500 μs | >2500 s | N/A | [66] |
| Ag/GO/FTO       | Defect migration Graphene: insulating matrix | LTP/LTD/STDP | 1.2 V/−1.2 V | 22−34 | 1 ms/1 ms | $10^8$ | $P_{set} = 0.045$ mW | [49] |
| Ni/GO/graphene  | Vacancy migration Graphene: insulating matrix | STP/LTP/STDP | N/A | N/A | N/A | N/A | N/A | [50] |
| Ti-Au/MoS$_2$/graphene/Ti-Au | Defect migration Graphene: insulating matrix | LTP/LTD | 3 V/−4 V | N/A | 100 ns/100 ns | $10^3$ s | N/A | [6c] |
| Ni-Au/MoS$_2$/graphene/Ni-Au | Charge trapping Graphene: insulating matrix | STP/LTP/STDP | 0.8 V/−0.8 V | N/A | 50 ns/50 ns | N/A | 37 fJ | [7b] |
| Ta/Ta$_2$O$_5$/AlN/graphene | Vacancy migration Graphene: back electrode | STP/LTP/STDP | 0.15 V/0.4 V | N/A | N/A | N/A | 0.01−1 fJ | [7a] |
| Al/AlO$_x$/graphene | Vacancy migration Graphene: back electrode | SET/DT transport | 1.5 V/−0.4 V | N/A | N/A | N/A | $P_{set} = 16.7$ nW | [7c] |
| Au/SnSe/graphene | Charge trapping Graphene: back electrode | STP/LTP/LTD/STDP | 0.8 V/−0.8 V | N/A | 100 μs/100 μs | N/A | 0.045 mW | [49] |
| RGO/GO-NCQD/graphene | Filament formation Graphene: back electrode | SET/STDP/SRDP/STDP | 0.8 V/−0.8 V | N/A | 10$^5$ | N/A | N/A | [51a] |

Synaptic graphene transistors can perform concurrent learning which updates synaptic weight without interrupting the signal transmission process. Both behaviors of excitatory and inhibitory synapses could be realized in one device because of the ambipolar conductance of the graphene channel. However, these transistors still have many shortcomings (Table 1): 1) low on/off ratio, especially compared with traditional transistors and synaptic memristors; 2) short retention time, indicating limited memory; 3) large device size, unavailable for large-area integrations where more than ten transistors are required to mimic one synapse; and 4) high energy consumption, much higher than that of the biological synapse (1−10 pJ). Graphene based synaptic memristors have the advantage of scalability, high on/off ratio, quick switching speed, low energy consumption, and continuously adjustable characteristic between LRS and HRS (Table 2). However, the simultaneous implementation of signal transmission and self-learning in memristors is challenging. The stability of these memristors under extreme conditions (e.g., high temperature, curved substrates) is very important due to their special memristive mechanisms which depend on the movement of ions, carriers, and defects. In addition, the following key subjects in synaptic transistors and memristors are essential: 1) dynamic synapses are important to mimic the synaptic plasticity in biology in higher dimensions to revolutionize the computation circuits; 2) different with most electrically driven ASs, photonic synapses integrating photosensitivity and information processing are critical to developing artificial visual perception systems; 3) flexible and biocompatible synapses should be realized to build efficient HMI.

In short, graphene-incorporated AI has made considerable progress in both software (ML) and hardware (ASs), but much remains to be done to achieve true AI in the future.

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Conflict of Interest
The authors declare no conflict of interest.

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artificial intelligence, graphene, machine learning, neuromorphic computing, synaptic devices

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