

# Introduction: Neuromorphic Materials

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The explosive growth in data collection and the need to process it efficiently, as well as the desire to automate increasingly complex tasks in transportation, medical care, manufacturing, security and many other fields have motivated a growing interest in neuromorphic computing.<sup>1</sup> Unlike the binary, transistor-based ON/OFF logic gates and separate logic and memory functionalities employed in digital computing, neuromorphic computing is inspired by animal brains that use interconnected synapses and neurons to perform processing, storage and transmission of information at the same location, while only consuming ~20 W or less of power. Motivated by the brain's efficiency, adaptability, self-learning and resiliency qualities, neuromorphic computing can be broadly defined as an approach to processing and storing information using hardware and algorithms inspired by models of biological neural systems. Present research in neuromorphic computing encompasses approaches that vary significantly in their degree of neuro-inspiration, from systems that only incorporate features such as asynchronous, event-driven operation or use crossbar arrays of non-volatile memory (NVM) elements to accelerate deep neural networks (DNNs), to designs that embrace the extreme parallelism, sparsity, reconfigurability, adaptability, complexity and stochasticity observed in nervous systems.<sup>2</sup> The term 'neuromorphic' computing is often credited to Carver Mead, who in the 1980s investigated Si-based analog electronics to replicate functions of the animal retina.<sup>3</sup> Earlier important advances in this field include the work of Frank Rosenblatt,<sup>4</sup> who proposed the concept of the perceptron, Bernard Widrow,<sup>5</sup> who used this concept to build one of the first analog neural networks, the Adaline and many other researchers (see ref. <sup>6</sup> for an historical perspective on neuromorphic computing). With the recent increase in the use of artificial intelligence and large language models, and rising concerns over the associated energy costs, interest in neuromorphic hardware has expanded rapidly. According to some estimates, driven largely by the drastic growth in the training use of artificial intelligence (AI) models using the current computing architectures, the energy cost of computing is projected to reach the energy supply worldwide by 2045.<sup>7</sup> While this is not a realistic outcome, it means that, if more efficient computing technologies are not developed -- soon -- the world will soon become one where demand for energy and market constraints limit the continued increase of societal access to AI and cloud services from data centers. Data centers used for training and use of these models consume hundreds of terawatt hours of electricity, already past 4% of the US electricity demand.<sup>8</sup>

Numerous established microelectronics manufacturers and startups have announced efforts to commercialize energy-efficient neuromorphic chips, with some systems that contain over one billion neurons, capable of supporting spiking algorithms, event-driven asynchronous communications, and some level of reconfigurability.<sup>1,9</sup> Nevertheless, the computational abilities of these schemes remain restricted to relatively narrow tasks and fall far short in terms of learning efficiency, contextualization and other aspects of general intelligence associated with mammalian brains<sup>10</sup>. In fact, the gap in computational abilities between artificial and biological systems with regards to general intelligence is enormous, despite very impressive progress in neuromorphic device technologies. To narrow this gap and to increase functionality and efficiency, a growing number of researchers have focused on exploring new neuromorphic device concepts that exploit spin, ionic, ferroelectric, microstructural, Mott, and other physical/chemical mechanisms to develop novel computational primitives for neuromorphic computing.<sup>11</sup> Many of these approaches have shown encouraging results for training and inference acceleration of deep neural networks,

edge processing of sensor signals, Bayesian neural networks, graph neural networks and physical reservoir computing schemes.<sup>12</sup> Also promising are approaches that explore coupling between different effects or state variables (e.g., Joule heating leading to Mott or spin transitions) to emulate complex neuronal dynamics, axon-like signal transmission and ensemble effects.<sup>13, 14</sup>

However, despite a growing number of compelling demonstrations of performance at the individual device level, the realization of practical neuromorphic computing systems based on emerging device concepts that can challenge digital Si CMOS-based computing systems remains a challenge. This is in part because most practical computing applications require scaling to many devices, as well as their integration with other components, including digital CMOS. Without such scaling and integration, validation of predicted computing advantages is difficult. Also difficult is the design of novel architectures and neuromorphic algorithms, which require a substantial level of abstraction at the device and small circuit scale, as well as a ‘user-friendly’ interface for programming and software development. The need to reliably fabricate at scale and integrate devices necessitates a detailed mechanistic understanding of the physical and chemical processes that underpin the computation primitives, the effects of material composition, structure, defects, interfaces, device geometries and dimensions, as well as external variables and drivers such as temperature and potential. This is a daunting task that calls out for a multidisciplinary codesign approach with contributions from chemistry, physics, materials science, electrical engineering, computer science and neuroscience.

In this thematic issue of Chemical Reviews, we include contributions from leading researchers engaged in advancing neuromorphic computing by focusing on the materials used to make neuromorphic hardware, the special mechanisms that enable computational primitives, their advantages in terms of efficiency and latency, and the challenges to making these new computing paradigms broadly applicable. The authors in this issue covered several distinct topics with some overlaps, that can be broadly categorized by the type of materials (e.g., organic versus inorganic) as well as applications (e.g., bio-integration versus chip scale systems). For example, S. Ramanathan et al. discuss how doping with protons of various organic and inorganic functional materials leads to behaviors useful for neuromorphic computing, and how these characteristics are related to biological neurotransmitters. They also discuss extensively the approaches and challenges to characterizing proton transport and effects in materials. Y. Zhou et al. review the scientific basis, status and challenges related to flexible neuromorphic materials and devices, including quantum dots, nanowires, nanocrystals, 2D layered semiconductors, nanomaterials (zero-, one-, and two-dimensional nanomaterials, and heterostructures), graphene and polymers. T-W. Lee et al. focus on biocompatible neuromorphic materials and devices, emphasizing both the sensor and the processing aspects involved in realizing functional interfaces between machines and the nervous system, including brain-computer interfaces and artificial muscle systems. V. K. Sangwan and M. C. Hersam et al., review the recent advances in 2D materials such as the transition metal dichalcogenides for neuromorphic hardware, with emphasis on establishing robust relations between the growth, fabrication, transport and device characteristics, as well as the challenges for integration of 2D materials and van der Waals heterojunctions for neuromorphic electronic and optoelectronic devices, and circuits. J. J. Yang et al. provide a detailed review of memristive devices that exploit ion dynamics to realize various characteristics useful for neuromorphic computing, ranging from analog synaptic behavior to complex dynamics that emulate neuronal models and involve coupling of several mechanisms. S. Kumar et al., review the history, mechanisms and opportunities for neuromorphic device engineering based on filament formation in devices based on various materials and configurations. They discuss both thermodynamic and kinetic aspects to provide a more unified understanding of the various phenomena and how these can be leveraged for advancing neuromorphic device concepts. A. A. Talin, Y. Li and B. Yildiz et al. review the scientific foundations and device applications of electrochemical random access memory (ECRAM), including extensive discussions of protonic, lithium-ion and oxygen vacancy types of electrochemical memories, their respective advantages and disadvantages, and the opportunities for realizing artificial synaptic and neuronal devices. D. Ielmini and G. Pedretti review the potential of resistive-switching random-access memory (RRAM) for in-memory

computing (IMC), outlining its advantages, and addressing the paths to address the requirements for a range of storage and computing applications, from materials, device, circuit, and application viewpoints. G. S. Syed et al. review the current state of phase-change materials (PCM), PCM device physics, and the design and fabrication of PCM-based chips for in memory computing and provide an overview of the landscape for applications and future developments.

We hope that these Reviews will help investigators interested in contributing to this rapidly evolving and fertile field get an appreciation of how the different aspects and challenges are connected and to identify opportunities for innovative solutions guided by fundamental understanding.

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