Additive manufacture of polymeric organometallic ferroelectric diodes (POMFeDs) for structural neuromorphic hardware

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ABSTRACT

Hardware design and implementation for online machine learning applications is complicated by a number of facets of conventional artificial neural networks (ANN), e.g. deep neural networks (DNNs), such as reliance on atemporal locality, offline learning using large datasets, potential difficulties in transfer from model to substrates, and issues with processing of noisy sensory data using energy-efficient and asynchronous information processing modalities. Analog or mixed-signal spiking neural networks (SNNs) have promise for lower power, temporally localised, and stimuli selective sensing and inference but are difficult fabricate at low cost. Investigation of beyond-CMOS alternative organic substrates may be worthwhile for development of unconventional neuromorphic hardware with pseudo-spiking dynamics for structural electronics integration in bio-signal processing and robotics. Here, polymeric organometallic ferroelectric diodes (POMFeDs) are introduced for development of printable ferroelectric in-sensor SNNs.

CCS CONCEPTS

• **Hardware → Flexible and printable circuits**; **Neural systems**; **Sound-based input / output**.

KEYWORDS

Event based spiking auditory sensors, spiking neural networks, ferroelectrics, additive manufacture.

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1 INTRODUCTION

Spiking neural networks (SNNs) represent a promising approach to address issues in the use of conventional artificial neural networks (ANNs), e.g. deep neural networks (DNNs), in online learning due to the potential for increased energy efficiency and better suitability for processing of spatiotemporal learning tasks including auditory sensing and processing [10] [36]. Implementations of SNNs vary from analog, digital, and mixed signal systems. Mixed signal and digital implementations offer the greatest flexibility yet have trade offs in terms of energy efficiency [7]. The promise of these neuromorphic implementations of spike based learning of spatiotemporal data are held back by lack of suitable sensing methods for generation of spiking inputs as well as high cost compute-in-memory hardware for low power and asynchronous processing of information. To take full advantage of the energy efficiency of SNNs, field programmable gate arrays (FPGAs) or neuromorphic processors should be connected directly to event driven sensors. Only a relatively small number of sensory devices have been developed to directly provide event driven data. The most prominent examples of this type of sensor have been in visual domains such as the dynamic vision sensor (DVS) used in applications such as tracking and motion detection [3] and event based tactile sensing for texture recognition [4]. In auditory devices both the silicon cochlea [6] and neuromorphic auditory sensor (NAS) [17] type devices have been developed. With implementation of the latter in auditory perception hardware for humanoid robots [12]. However, FPGA based technologies may not easily be designed into wearable applications especially those with structural or implantable electronics requirements such as in cochlear implants. The alternative option, namely specialised neuromorphic hardware, remains expensive for device implementation with access via cloud servers for applications requiring online learning of sensory input still in development. More generally, the cost of fabrication of many complementary metal-oxide-semiconductor (CMOS) based artificial neuromorphic systems mean that in-sensor implementations can be difficult to develop in particular in situations where a rapid prototyping approach is preferred.

Alternative approaches using materials that exhibit reversible switching in response to diverse stimuli have been studied as a way to integrate pseudo-synatic responses directly into the operation of the sensor. Of particular relevance are approaches that utilise in-sensor resistive switching properties of devices such as memristors to perform memory and computational functions co-incidentally in the same volume as a sensing element. Much of this work builds on initial exploratory research in the 1950s [27]. Recent research efforts in this regard include modulation of the electrical [35] or optical property [32] of the resistive switching layer on application of an external electric field, magnetic field, or light in response to external stimuli. These stimuli selective sensors with in-built memory functionality have the potential to function as low power and asynchronous systems suitable for realisation of event based

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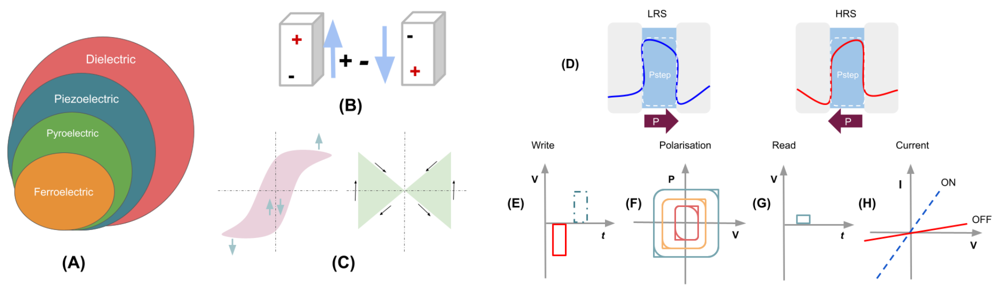

Figure 1: (A) Illustration of the relationship between ferroelectric, piezoelectric, pyroelectric and dielectric phenomena;(B) A basic illustration of ferroelectric polarisation in response to external electrical field;(C) generic I-V and G-V hysteresis curves for ferroelectrics;(D) illustraion of pstep based incremental low resistance and high resistance states;(E) pulse programming;(F) multiple frequency incremental polarisation;(G) read signal;(H) on/off current.

physical intelligence such as those used in bio-signal matched neuroprothetics [8] or in development of relevant control methods in robotics [2].

Ferroelectricity, and its appearance in biological systems alongside concurrent effects such as piezoelectricity represent an underexplored aspect of methods to establish principles for design and engineering of pseudo-synaptic elements and in-sensor memory for neuromorphic devices. Ferroelectric devices, including those based on mechanically flexible materials such as polymers, are a promising basis for exploration of alternative neuromorphic substrates for in-sensor machine hearing and auditory learning applications based on ferroelectric polarisation, i.e. the "ferroelectric plasticity" that can be obtained by regulating the amplitude or duration of the applied voltage pulses. As outlined in figure 1, this phenomena can be used in development of resistance switching devices (RSDs) based on the incremental polarisation with concurrent low voltage sensing or "read" functions. The two-terminal type ferroelectric artificial synapses comprises of two electrodes that are separated by a film. When a pre-synaptic signal is applied to one electrode, an update of the conductance is generated accordingly, and the synaptic weight can be readout from the other electrode (see description in figure 2). In this way, spike signals are transferred from pre-synaptic terminal to the post-synaptic one. Ferroelectrics also benefit from high potential for realising in-sensor memory in auditory sensors as a result of co-location of piezoelectric phenomena. This makes ferroelectric synapses a promising platform for realisation of in-sensory spike based learning of spatiotemporal data such as vibrotactile or auditory stimulus. Polymeric ferroelectrics offer many advantages for these applications with trade offs in terms of their potential lack of integration into existing manufacturing processes such as CMOS. However, while they are comparatively underdeveloped solution processing methods for organic ferroelectric polymers allow for greater access to rapid prototyping and lower cost fabrication.

In this paper, a polymeric ferroelectric synapse based on an organometallic glycinate complex is introduced and a method for additive manufacture is demonstrated based on polymeric organometallic ferroelectric diodes (POMFeDs). The basic ferroelectric synapse properties are demonstrated including hystersis I-V and G-V curves, cycle-to-cycle performance, and in-sensor co-location of ferroelectric polarisation programming in response to stimuli. These results provide a proof of concept for this new ferroelectric diode-type synaptic sensor combined with early identification of limitations of our fabrication approach and performance of the diode.

2 RELATED WORK

In conventional hardware systems, sensors are typically externally connected to memory and processing units via the Von Neumann computing architecture. Sensing occurs in the analog domain, which leads to the generation of large amounts of raw data from the sensor. The large amounts of redundant analog data, including unnecessary background noise and related phenomena, must be converted to digital data via an analog-digital-converter (ADC), then transferred to processing units or cloud-based computing systems. As a result the sensory data processing requires data communication that is energy inefficient because of the transfer of information from sensors to the memory element. This conversion and transportation causes significant issues in terms of energy consumption, delayed response time, and the communication bandwidth, all of which are important for machine hearing applications with strict delay and power-consumption requirements. Therefore, this data transportation bottleneck issue in integrated sensor and processor systems should be mitigated for fast and efficient information processing.

In-sensor memory systems refer to the incorporation of sensing and memory elements within a single volume. The main advantage of in-sensory memory is in reduction of data transfer times and footprint size as a result of both elements being co-located within the same material volume within the footprint. In artificial auditory and vibrotactile devices, this process can be replicated in part by voltage driven modulation of the electrical resistance, namely resistance switching, in response to external stimulus. Resistance switching (RS) was explicitly identified in the 1960s in binary oxides [14]. Since then many methods and material combinations have been suggested for use as the resistive switching layer in neuromorphic substrates including ion migration materials [18], phase change materials [5], conducting filaments [37], and ferroelectrics [26]. Stochastic processes including the formation of conducting filaments and phase change have been shown to change morphology

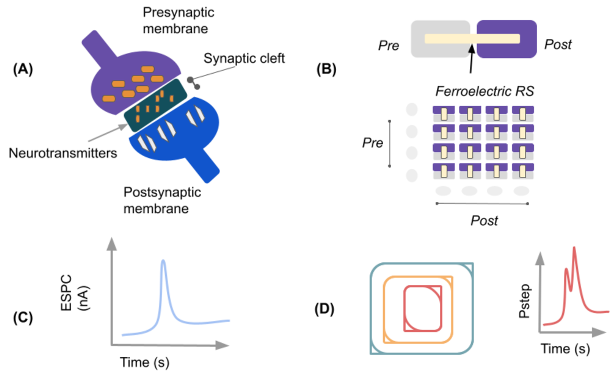

Figure 2: (A) Illustration of pre-and-post synaptic connections;(B) schematic neuromorphic representation of pre-and-post synaptic connections based on the electrode-ferroelectric-electrode structure;(C) excitatory post-synaptic current (ESPC);(D) ferroelectric polarisation (pstep) and corresponding current.

cycle to cycle leading to variability in the RS behaviour that may be disadvantageous [24]. Interface-type RS layers,which includes ferroelectrics, offer advantages in spatially and temporally localised learning relating to superior stability, more predictable stochastic and hysteretic cycle-to-cycle parameters, non-linear current with self-rectification characteristics and area scalability [24].

Ferroelectricity was discovered in 1920 when J. Valasek demonstrated that the spontaneous polarization of Rochelle salt could be macroscopically inverted by an applied external electric field [34]. Since then ferroelectrics have attracted attention due to concurrent phenomena including pyroelectricity, piezoelectricity, non-linear optical properties and inverse piezoelectricity [24]. Depending on the application requirements ferroelectric materials can serve as versatile transducers facilitating sensing, actuation, data storage, energy harvesting, electro-optic devices, amongst other technical uses [31]. Various ferroelectric devices have been explored for Boolean memory devices because of their bistable remanent polarisation response upon application of electric fields. This enables digital 0 and 1 states to be programmed based on the state of the remanent polarisation +Pr and -Pr respectively [31]. Remanent polarisation states can also be programmed to intermediate values between the -P and +P limits. This quality is especially attractive for artificial synapses and devices with the time stability of the intermediate polarisation states in ferroelectrics being advantageous [31]. Ferroelectric devices based on capacitors, transistors and tunnel junctions can realise artificial neurons or synapses but face many implementation challenges. Ferroelectric synapses demonstrating spike-timing-dependent-plasticity (STDP) have been reported using ferroelectric field-effect transistors (FeFETs) [28] and ferroelectric tunnel junctions (FTJs) [19]. FeFETs are comprised of a ferroelectric capacitor as gate insulator, modulating the transistors threshold voltage that can be sensing non-destructively by measuring the drain-source current. FTJs feature a ferroelectric layer sandwiched between two electrodes thus modifying the tunnelling electro-resistance. A polarization-dependent current is measured non-destructively with a read pulse. Despite their promise, the adaptation of existing ferroelectric devices to neuromorphic substrates has many issues. Capacitive ferroelectric synapses require accessing the intermediate polarisation states via applying an external field and thereby altering the polarisation state. This readout method is destructive as the polarisation state must be restored after every readout operation. Alternative design methods such as the use of a second reference capacitor increases the complexity. FeFETs have issues with technological implementation, limited scalability and data retention issues.

Ferroelectric diodes (FeDs) are underexplored for neuromorphic applications in terms of structural integration as part of devices suitable for use cases such as some printable electronics and in bioelectronics. The characteristics of FeDs are centred on spontaneous polarization that can be inverted by an external electric field, resulting in a hysteresis loop. The existence of the spontaneous polarisation results in the formation of positive and negative bound polarization charge at the corresponding polarized surfaces [31]. The diode type device is advantageous for resistance switching as a result of its bistability only at a forward bias. The rectifying behavior of the diodes can eliminate crosstalk and allows for the realization of a 2D array without the need for additional addressing or driving devices [31]. Ferroelectric diodes also have wider scope for implementation of multi-frequency resistance programming in the same individual element.

3 DESIGN AND FABRICATION OF THE POMFED

Inorganic ferroelectric oxides have previously been used in ferroelectric synapses. However, poor mechanical endurance makes them non-biocompatible and stiffness characteristics and complex fabrication mean they cannot meet the requirement for area-scalable applications. In contrast, ferroelectric polymers have excellent flexibility and intrinsic analog switching characteristics [24]. In terms of fabrication, inorganic ferroelectrics require procedures that are highly sensitive to initial conditions such epitaxial growth and complex crystal growth optimisations such as use of specific substrates for lattice matching. In contrast, polymeric ferroelectric materials can be easily dissolved in an organic solution and directly printed onto most flexible substrates. This means that their easy processing, low cost, and flexibility offer feasible solutions for large scale design of synaptic elements and network formation while also having excellent properties for use in bio-informational interfaces such as those required in auditory and vibrotactile learning devices due to their bio-compatibility.

The commonly used ferroelectric polymer, poly(vinylidene fluoride) (PVDF) and its copolymers offers better ferroelectricity and thermal stability over other counterparts, such as nylon [1], polyurea [21], croconic acid [15], liquid-crystals [9], and poly(m-xylylene adipamide) (MXD6) [23]. A variety of methods for fabrication of ferroelectric polymer films including PVDF have been proposed including spin coating [29], Langmuir–Blodgett coating [38], additive manufacturing [16]. However, there are a number of performance limitations with PVDF including strong temperature dependence in its piezoelectric and ferroelectric properties requiring careful calibration and temperature compensation techniques. Other issues include the polarisation stability and relatively narrow frequency response. Fabrication drawbacks to the use of PVDF including high cost, high dissipation factor, availability, and issues relating to bio-compatibility and sustainability (e.g. non-biodegradable)[24]. Organometallics, including metallic complexes of bio-polymers such as glycine, have the potential to offer advantages over conventional approaches to polymer ferroelectrics due to the possiblity of improved ferroelectric and piezoelectric response, tunable thermal stability, and ability to engineer materials with suitable bio-compatibility and degradation profiles [11].

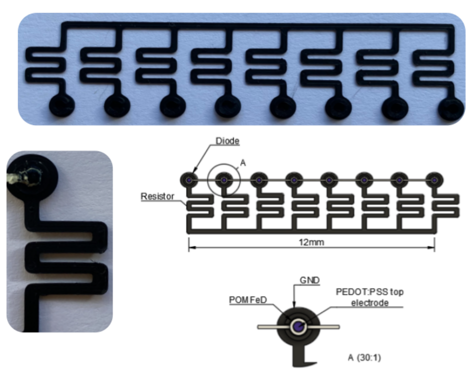

Figure 3: Design overview and 3D printed structure including inbuilt resistor.

3.1 Fabrication approach

The PVA filament (eSun) for the ferroelectric layer was coated with the organometallic glycinate solution and printed using an FDM printer (Original Prusa MINI+, Prusa) at temperatures not exceeding 180C°. The top electrode layer was 3D printed using a conductive carbon filament (ProtoPasta). The diode structure was completed with a top coating of PEDOT:PSS (Clevios PH 1000) deposited using a robot arm fitted with a custom ink depositing end effector. The total footprint length of the smallest successfully printed multiple diode device including was 12mm containing 8 diodes in total. Actual photos of the device and diagrams can be seen in figure 3.

4 BASIC CHARACTERISATION OF THE POMFED

Following printing the POMFeD was characterised to establish its suitability for use in structural neuromorphic hardware.The hysteresis experiment is based on driving the ferroelectric diode element in series with a $2.2k\Omega$ printed resistor structure. This method demonstrates the response as a time series, current-vs-voltage (I-V) or conductance-vs-voltage (G-V) plot. Tests were carried out at multiple signal frequency showing the characteristic ferroelectric hysteresis with some issues relating to voltage drop across the device at lower frequencies outlined in figure 4. Longer cycling of the hysteretic response was investigated for a single frequency over 100k cycles (see figure 5).

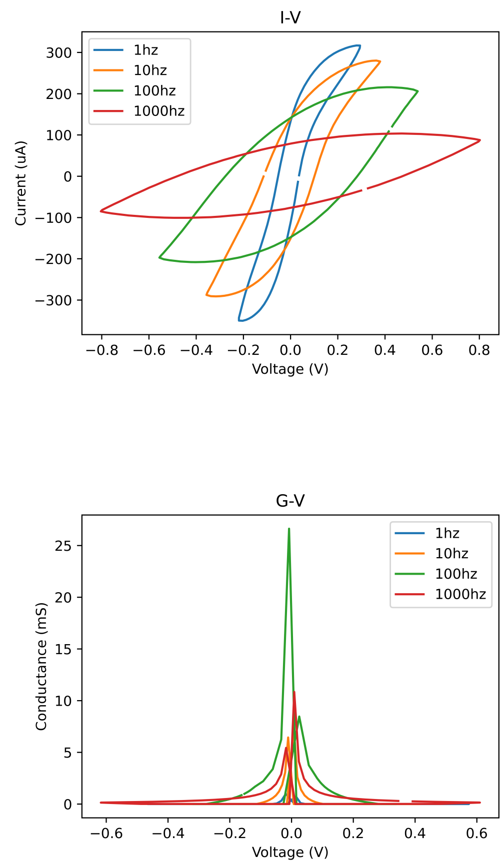

Figure 4: I-V (top) and G-V (bottom) curves for the 3D printed POMFeD showing ferroelectric voltage/current hysteresis loops at different frequencies and conductance change at different frequencies as a function of voltage respectively.

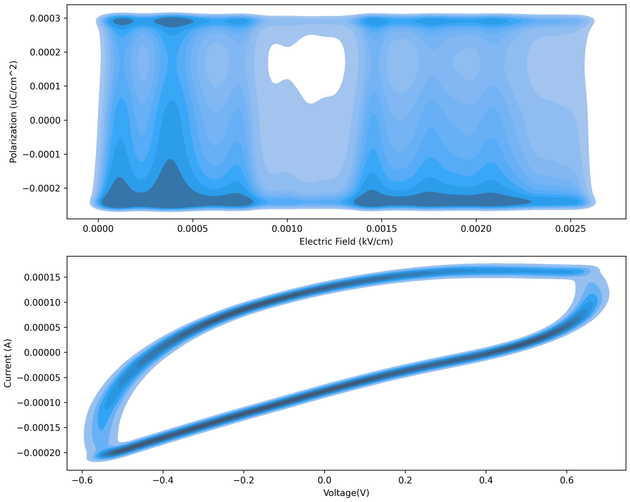

Figure 5: Current-polarisation versus electric field and I-V for single frequency 1kHz cycling of the device over 100,000 cycles at 1V.

The post-pulse response experiment is set up similarly with ferroelectric diode element in series with the in-built resistor. One or more pulse waveforms were applied and the instantaneous and post-pulse responses were recorded. The instantaneous response was recorded as a time series of voltage (1+,2+ vs T) and current (I vs T). The instantaneous pulse response for each following 'read pulse' was also recorded and updated after every read pulse. The resistance programmability of the device shows increasing and decreasing current in response to positive and negative voltages. This indicates the suitability of the device for implementation of basic STDP behaviour such as long-term depression (LTD) and long-term potentiation (LTP).

5 STDP AND SRDP BEHAVIOUR

The general characteristics of the device including ferroelectric response, conductance/resistance programming based on the polarisation, and the pulse-programming are now outlined. The application of the off-to-on (LTP) using positive pulses and the on-to-off (LTD) for negative switching pulses. The results for current transition show a basis for development of pseudo-synaptic behaviours such as LTD and LTP as illustrated in figure 6 and shown via pulse programming tests and cumulative weights in figure 7.

The experiment applied pulses with a fixed width of 100 μs and an amplitude of 1V to produce the LTP-LTD type behaviours (top) and resulting accumulative weights (below). This stimuli-responsive behaviour shows pseudo-synaptic ESPC and ISPC responses based on the incremental polarisation or alternatively the current/conductance values as illustrated in figure 6 (A) and (B). When combined with event based input such as from suitably designed sensors the approach can achieve memory functionality based on the remanent polarisation response and non-destructive "read" voltage as described in figure 6 (D) and (E).

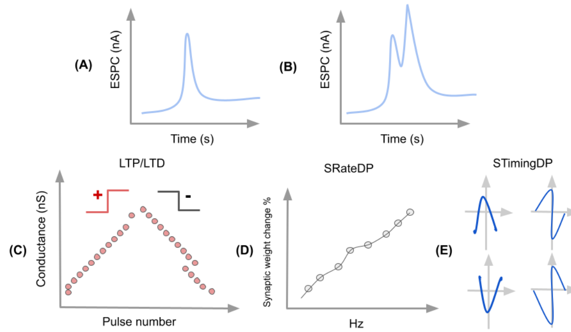

Figure 6: (A) excitatory post-synaptic current (ESPC);(B) inhibitory post-synaptic current (ISPC);(C) Long term potentiation and depression (LTP/LTD) ;(D) Spike rate dependent plasticity (SRDP);(E) spike timing dependent plasticity (STDP).

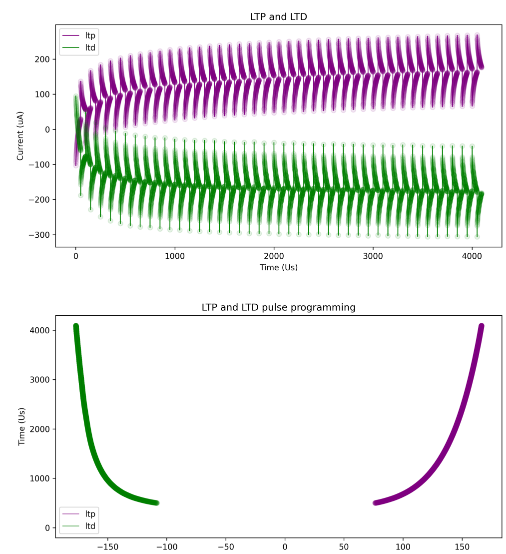

Figure 7: LTP-LTD type behaviours (top) and cumulative weights (bottom).

More specifically, the device polarisation is assumed to work on the basis of the application of a bias just below the coercive bias of the RS material which results in the nucleation of isolated domains with reversed polarisation. When the bias is removed, e.g. via the voltage/time dynamics of the pulse programming, the nucleated domain cannot retain its polarisation state due to insufficient energy. The domain loses its reversed polarisation and returns to its original polarisation state. Multiple applications of pulses can prevent this reversal for the nucleated domains and thus produce LTD-LTP behaviours depending on the stimulus parameters [31]. However, this is a complex material phenomena and relies on the time interval of the pulses to be shorter than the polarisation relaxation dynamics.

6 IN-SENSOR MEMORY

One final aspect of the diode device is the concurrent ferroelectric/piezoelectric response of the organometallic glycine RS layer. This property of the material can be used for applications such as event based auditory sensors in combination with spike based information encoding and learning in a single substrate material as outlined in figure 8. The concept was tested for establishing a proof of concept using two approaches: (a) applied voltage stimulus (1V at variable 20hz-20kHz frequency range in 2s pulses) (figure 9) and (b) at zero voltage input, corresponding to zero current-polarisation of the element, to demonstrate the potential for event based sensing (figure 10). The voltage stimulus experiment shows hysteresis loops corresponding to the various frequencies of the input stimulus resulting in ferroelectric polarisation vs the electric field. The zero polarisation experimental results show the piezoelectric response of the material and corresponding polarisation values at voltage input levels produced by the piezoelectric response only. This shows a very basic demonstration of event based sensing with representative SRDP and STDP output suitable for further development.

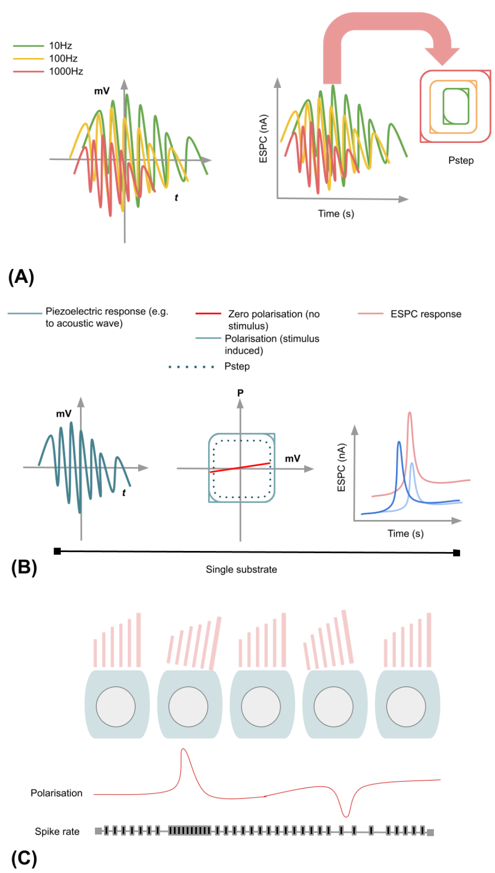

Figure 8: (A) Conceptual illustration of the piezoelectric signal input in response to audio and co-located ferroelectric polarisation;(B) co-located sensory-memory function where a input signal at a certain frequency causes polarisation from the zero polarisation state which can be represented as changing ESPC values within a neuromorphic context;(C) Speculative biological analogy with the polarisation dynamics of the stereocillia.

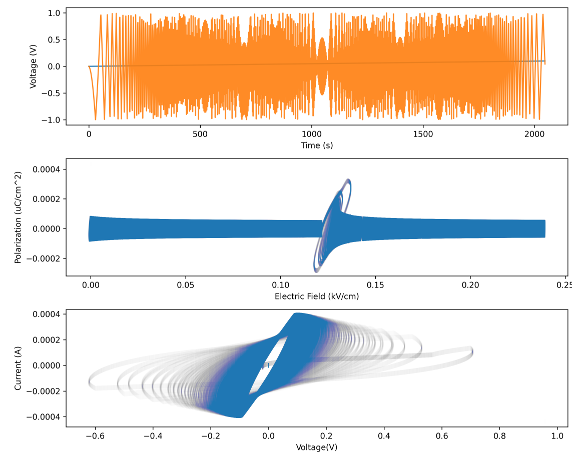

Figure 9: Top to bottom: repeated input voltage sweep (20hz-20kHz) stimulus (top); polarisation values; hysteresis curve (bottom).

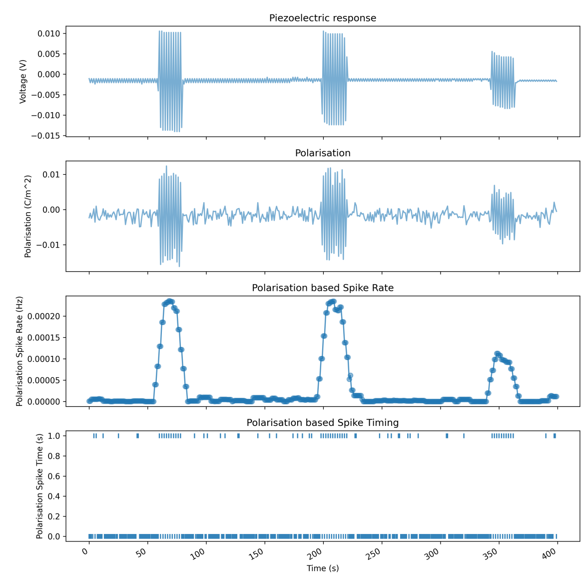

Figure 10: Top to bottom: piezoelectric response of the material from a zero polarisation or resting state (i.e. no power input); polarisation; representative spike rate frequency encoding; representative spike timing encoding.

The zero polarisation response could be used to develop pseudo-synaptic responses based on mechano-transduction in the inner hair cell (IHC) of the inner ear (see figure 8 (C)). In the IHC the position of the hair cells relative to one another based on the sound pressure in the cochlea dictates the electromechanical properties of the resulting spikes including polarity. In the artificial device the co-located piezoelectric and ferroelectric effects can be combined to couple immediate transducer response with incremental polarisation states.

7 DISCUSSION AND FUTURE WORK

Additive manufacture is a potentially useful approach for fabrication of certain types of neuromorphic substrates based on organic or organometallic materials as has been shown previously for organic electrochemical transistors (OECTs) [20]. While unconventional, there are examples in the literature of 3D printed features with nanoscale resolution that utilise approaches such as electropolymerisation in combination with deposition in 3D [13]. A wide range of alternative and beyond-CMOS type manufacturing techniques may prove advantageous when working with organic or organometallic ferroelectric polymers as well as other organic and biological material substrates. More generally, evidence of pseudo-spiking behaviours in a wide range of organic and biological substrates including stimuli-responsive and oscillatory pseudo-spiking in protenoid computing substrates (which share many similarities with organometallic amino acid polymers) [22] necessitates the development of beyond-CMOS fabricaiton methods. Organic substrates may prove to be useful in certain applications when coupled with additive fabrication methods and online learning algorithms such as reservoir computing [33], phasor [25] and oscillatory neural networks [30]. Further, many applications that would benefit from bio-compatible and bio-integratable elements only require a small number of individual pseudo-synaptic neurons (e.g less than 100 elements). Coupled with multiple frequency operation the number of synaptic operations per element could be increased dramatically offering benefits for applications such as auditory sensors and auditory neurotechnologies where processing of frequency input ranges typical in human hearing of 20hz-20kHz in an online manner are important considerations. Structural design considerations also work in favour of the approach outlined here. In particular, for implanted devices where the patient-specific shape and mechanical properties of the overall device are compared to overall processing power, number of pseudo-synaptic elements and energy per synaptic event.

In terms of fabrication approach, the use of standard inexpensive 3D printers for fabrication of ferroelectric diodes based on organics is less explored in the literature and is unproven. However, the use of 3D printing for POMFeD based synaptic weights may be a promising approach for the highlighted applications where the structural integration of neuromorphic hardware into unconventional substrates is important. In addition, the approach allows for rapid prototyping of the overall device in a single volume or multiple interconnecting parts. Prototyping in this way lowers the barrier to innovation that is having a marked impact on progress in established machine learning fields such as DNNs. Direct comparisons with fabrication techniques for arrays organometallic glycinates are not possible as it has not previously been investigated in the literature on neuromorphic hardware. However, in general, there are a number of limitations to the approach taken in this paper including obvious issues relating to footprint size and resolution of the deposition when compared to inorganic ferroelectrics fabricated using CMOS, photolithography, electron-beam lithography and similar conventional methods. Other methods of additive manufacture such as electrodeposition, electropolymerisation, and sonocrystallisation have potential for higher resolution and smaller scale sizes and will be explored in future work [13]. Despite the need for future development, the approach outlined here is orders of magnitude lower cost than even the cheapest approach to CMOS and other conventional fabrication techniques for organic and inorganic ferroelectrics as it is solution processable, does not require careful optimisation of crystal growth and substrate lattice matching. The RS material is bio-compatible and bio-integratable and could form a platform for wearable, implantable, and semi-implantable computing substrates. The in-sensor SNN concept would be greatly improved through use of higher resolution and multimaterial based manufacture such as development of a printable MEMs piezoelectric/ferroelectric transducer with co-located synaptic weights. This will also be the subject of future work as the approach to do this with organometallic glycinates is not currently available to the best of our knowledge. Additional future work will also focus on integration of these described approaches with suitable online learning methods including those based on SRDP/STDP, reservoir computing, phasor or oscillatory neural networks. Finally, the use of concurrent piezoelectric/ferroelectric effect in organometallics may be interesting for further investigation of mixed signal acoustic and electronic circuit technologies with useful non-electronic hysteretic and transport effects.

8 CONCLUSION

A 3D printed POMFeD was demonstrated as a ferroelectric diode for neuromorphic applications. The diode shows early proof of concept of characteristics that are suitable for structural implementations of synaptic weights in applications of printable electronics and bio-electronics that utilise unconventional substrates. In addition, organometallic ferroelectric layer forms a contribution to use of bio-compatible ferroelectric polymers such as glycinates in future device designs. The overall approach prioritises the fabrication of low-cost in-sensor SNNs with suitability for integration into mixed-signal or analog learning methods.

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