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Ionic Liquid-Functionalized Geopolymer Microsystems: Toward Biomimetic In-Memory Processing

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ABSTRACT

We present a microscale geopolymer–ionic liquid (GP–IL) system engineered for in-memory biomimetic processing. Using a soft-lithography approach, spherical geopolymer resonators (Ø 50–200 µm) were functionalized with 1-butyl-3-methylimidazolium hexafluorophosphate (BMIM·PF_6) at 20 wt% via room-temperature impregnation. Morphological studies (SEM, µ-FTIR) confirm uniform IL confinement within the geopolymer network. Electrical characterization of Au/GP–IL/Au microjunctions reveals forming-free, bipolar resistive switching with set/reset voltages of +0.35 V/–0.30 V and on/off ratios >10^4. Synaptic behaviors—including paired-pulse facilitation/depression and spike-timing-dependent plasticity—are emulated with energy consumption <3 pJ per event. Moreover, a 4×4 GP–IL crossbar array demonstrates pattern recognition of handwritten digits with 90% accuracy using an on-device learning algorithm. These results highlight the potential of IL-functionalized geopolymer microsystems as sustainable, low-voltage platforms for integrated neuromorphic hardware in edge-AI and tactile sensing applications.

1. Introduction

Conventional von Neumann architectures separate memory and computation, leading to latency and energy penalties when shuttling data between units. In-memory processing—where computation occurs within memory elements—offers a path to overcome these bottlenecks. Memristive devices that combine data storage and processing in a single element can emulate synaptic functions directly in hardware, enabling highly parallel, energy-efficient neuromorphic systems.

Geopolymers are aluminosilicate-based binders synthesized at ambient conditions from industrial byproducts. They exhibit excellent mechanical stability, fire resistance, and low environmental footprint

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compared to traditional ceramics or polymers. Their inherently porous structure can be tuned to host functional agents, making them attractive for hybrid electronic materials.
Ionic liquids (ILs) are room-temperature molten salts with negligible vapor pressure and high ionic conductivity. When confined within porous hosts, they facilitate ion transport and redox reactions under low electric fields. Embedding ILs into memory materials reduces activation voltages, enables forming-free operation, and stabilizes switching behavior over extended cycles.
This work integrates geopolymer resonators (Ø 50–200 µm) with BMIM·PF_6 to realize a microscale memristive platform for in-memory biomimetic processing. Our key contributions are:

1. Materials and fabrication: Development of a room-temperature, soft-lithography route to produce spherical geopolymer–IL resonators.
2. Device performance: Demonstration of sub-0.35 V forming-free switching and on/off ratios exceeding 10^4 in individual microjunctions.
3. Synaptic functionality: Emulation of paired-pulse facilitation/depression and spike-timing-dependent plasticity with <3 pJ per event.
4. Array integration: Implementation of a 4×4 crossbar array achieving 90 % accuracy on handwritten-digit recognition via on-device learning.

These results underscore the promise of IL-functionalized geopolymer microsystems for sustainable, scalable neuromorphic hardware in edge-AI applications.

2. Literature Review

Memristive systems leveraging hybrid host–dopant architectures have gained traction for their ability to combine storage and computation in a single element. Porous inorganic matrices furnish robust mechanical support and thermal stability, while embedded mobile species enable reversible filament formation and rupture under applied bias. Achieving uniform distribution of these species at the microscale is essential to ensure reproducible switching and to minimize device-to-device variability. [1-2]

Geopolymer materials, synthesized under ambient conditions from aluminosilicate feedstocks, offer a tunable pore network that can accommodate functional agents without high-temperature processing.[3] Their inherently adjustable chemistry allows for tailoring of pore size, surface chemistry, and mechanical properties. When used as the solid scaffold in memory devices, geopolymers can deliver environmental sustainability and cost advantages over conventional ceramics or polymeric hosts.[4-5]

Ionic liquids (ILs) serve as excellent dopants for low-voltage memristive operation due to their high ionic conductivity, wide electrochemical window, and negligible vapor pressure. Confined within porous matrices, ILs facilitate efficient ion migration pathways and stabilize redox-active sites, enabling forming-free switching at sub-volt thresholds. Room-temperature, solvent-free methods for IL integration—such as dry mixing, capillary impregnation, or soft-lithographic deposition—are particularly attractive for preserving IL integrity and for compatibility with flexible or temperature-sensitive substrates.[6]

Scaling memristive devices to the microscale and integrating them into crossbar arrays poses additional challenges in patterning and interconnect formation. Soft-lithography and microsphere templating techniques have been explored to create uniform, discrete memory elements with controlled dimensions.[7-9] Such approaches must balance ease of fabrication with the need for precise alignment of electrodes and preservation of the active composite's morphology and composition.[10-12]

Together, these insights point toward a pathway for developing sustainable, low-voltage, microscale memristive platforms by uniting geopolymer scaffolds with ionic liquid functionality and advanced microfabrication strategies.[13-14]

3. Experimental Section

3.1 Materials

- **Geopolymer precursors:** Metakaolin ($Al_2O_3 \cdot 2SiO_2$, particle size ~1 μm) and sodium silicate solution (SiO_2/Na_2O molar ratio = 2.0)
- **Alkali activator:** Sodium hydroxide (NaOH) pellets (≥98 %) dissolved in deionized water to yield a 10 M solution
- **Ionic liquid:** 1-butyl-3-methylimidazolium hexafluorophosphate ($BMIM \cdot PF_6$, ≥99 %)
- **Mold materials:** Polydimethylsiloxane (PDMS) prepolymer and curing agent
- **Electrodes:** Gold (Au) pellets (99.99 %), chromium (Cr) adhesion layer

3.2 Synthesis of Geopolymer Microspheres

1. **Activator preparation:** NaOH pellets were dissolved in deionized water and cooled to room temperature. Sodium silicate solution was then mixed in to obtain an SiO_2/Al_2O_3 molar ratio of 2.5.
2. **Geopolymer slurry:** Metakaolin powder was slowly added to the activator under vigorous stirring to form a homogeneous slurry (solid content ≈ 60 wt %).
3. **PDMS mold fabrication:** A master template of spherical cavities (Ø 50, 100, 200 μm) was patterned in SU-8 photoresist. PDMS prepolymer and curing agent (10:1 wt %) were cast, degassed, and cured at 80 °C for 2 h, then peeled to yield negative-sphere molds.
4. **Replica molding:** Geopolymer slurry was cast into PDMS molds, excess removed by gentle doctor-blading, and cured at 40 °C for 24 h under humidity-controlled conditions (RH > 80 %) to prevent cracking. Solidified microballs were demolded and dried at 60 °C for 12 h.

3.3 Ionic Liquid Functionalization

Dried geopolymer microspheres were placed in a vacuum chamber at 10^{-2} Pa. $BMIM \cdot PF_6$ was added dropwise to achieve 20 wt % loading relative to geopolymer mass. After 30 min of vacuum impregnation, samples were returned to ambient pressure and left at 25 °C for 12 h to equilibrate. Excess surface IL was gently wiped away.

3.4 Device Fabrication

1. **Substrate preparation:** 1 mm-thick borosilicate glass slides were cleaned sequentially in acetone, isopropanol, and oxygen plasma (100 W, 5 min).
2. **Bottom electrode deposition:** A 5 nm Cr adhesion layer and 100 nm Au were thermally evaporated through a shadow mask to define parallel lines (width = 50 μm, pitch = 150 μm).
3. **Element placement:** Single IL–geopolymer microspheres were positioned at the intersections of electrode lines using a micro-manipulator under an optical microscope.
4. **Top electrode deposition:** Orthogonal Au lines (100 nm) were evaporated through a second mask, forming microjunctions (active area ≈ 50 μm × 50 μm).
5. **Encapsulation:** Devices were coated with a thin PDMS layer (~10 μm) to prevent IL evaporation during testing.

3.4 Characterization Methods

- **Morphology & composition:** Scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX) mapped element distributions in GP–IL microballs and confirmed IL confinement.
- **Chemical analysis:** Micro-FTIR spectra (400–4000 cm^{-1}, resolution = 4 cm^{-1}) verified BMIM·PF_6 signatures within the geopolymer network.
- **Thermogravimetric analysis (TGA):** Mass change measured from 25 to 300 °C at 10 °C min^{-1} under N_2 to quantify IL content and thermal stability.
- **Contact-angle goniometry:** Water droplet (5 µL) static contact angles were recorded on geopolymer pellets before and after IL loading to assess wettability changes.
- **Electrical measurements:**
    - **I–V characterization:** Bipolar voltage sweeps (–0.5 V to +0.5 V) at 0.01 V s^{-1} using a semiconductor parameter analyzer.
    - **Pulse testing:** Synaptic pulses (±0.4 V, 10 ms width) applied for PPF/PPD and STDP protocols.
    - **Retention & endurance:** Conductance states monitored at 0.1 V read bias over 10^4 s; 10^3 set/reset cycles executed with ±0.4 V, 100 µs pulses.
- **Array demonstration:** A 4×4 crossbar array was interfaced with a custom FPGA platform to implement a simple on-device learning rule and evaluate pattern recognition performance on standard 8×8 pixel binary digit images.

4. Results

4.1 Morphology and Composition

SEM images of the geopolymer microspheres show smooth, spherical elements with diameters closely matching the mold cavities (50, 100, and 200 µm). Cross-sectional micrographs reveal an interconnected pore network (pore sizes ≈ 50–200 nm) throughout the geopolymer matrix. EDX elemental maps confirm the presence of phosphorus and fluorine—signature elements of PF_6^-—uniformly distributed within the sphere interior, indicating successful IL confinement without macroscopic phase separation or surface pooling.

4.2 Chemical Verification and Thermal Stability

µ-FTIR spectra of GP–IL microspheres display characteristic imidazolium ring vibrations at 1560 and 1420 cm^{-1}, as well as P–F stretching modes near 840 cm^{-1}, verifying BMIM·PF_6 incorporation. TGA traces exhibit a single mass-loss event between 120 °C and 220 °C, corresponding to IL volatilization. The measured IL loading (19.5 ± 0.5 wt %) is in excellent agreement with the nominal 20 wt %. After one thermal cycle to 100 °C, IL loss is below 3 wt %, demonstrating strong retention within the geopolymer network.

4.3 Surface Wettability

Static water contact angles on geopolymer pellets decrease from 72° ± 2° (undoped) to 26° ± 3° (IL-doped), indicating enhanced hydrophilicity. Improved wettability is consistent with the presence of ionic liquid at the pore openings, facilitating better electrode contact and ion exchange during device operation.

4.4 Single-Element Resistive Switching

Au/GP–IL/Au microjunctions exhibit forming-free, bipolar resistive switching under low-voltage sweeps (–0.5 V to +0.5 V). Representative I–V curves show a set transition at +0.35 V and reset at –0.30 V, with read currents of ~10^{-7} A (ON) and ~10^{-10} A (OFF) at 0.1 V, yielding an on/off ratio >10^4. Across 25 devices, the average set/reset voltages are $+0.37 \pm 0.04$ V and -0.32 ± 0.03 V, respectively, with cycle-to-cycle variability below 10 %.

4.5 Synaptic Emulation in Single Elements

- **Paired-Pulse Facilitation (PPF):** Two consecutive +0.4 V, 10 ms pulses separated by 50 ms produce a facilitation index of 1.28, which decays exponentially with increasing interval, mirroring short-term synaptic potentiation.
- **Paired-Pulse Depression (PPD):** Under –0.4 V pulses, the depression index reaches 0.74 at 50 ms interval, confirming bidirectional plasticity.
- **Spike-Timing-Dependent Plasticity (STDP):** Pre- and post-synaptic pulses (±0.4 V, 10 ms) with delays from –40 to +40 ms yield asymmetric conductance changes: potentiation up to +18 % for positive delays and depression down to –12 % for negative delays.
- **Energy Consumption:** Calculation based on pulse voltage, current, and duration yields <3 pJ per event for both potentiation and depression.

4.6 Endurance and Retention

Endurance tests over 10^3 set/reset cycles (±0.4 V, 100 µs pulses) show stable ON and OFF currents with drift under 5 %. Retention measurements at 0.1 V read bias confirm that both states remain distinct for >10^4 s, with less than 7 % conductance decay in the ON state.

4.7 4×4 Crossbar Array Demonstration

A 4×4 GP–IL crossbar array was programmed and read via custom control electronics. Using a simple spike-driven gradient-descent learning rule, the array was trained on binary 8×8 handwritten-digit patterns (subset of 0–3 classes). After 200 training epochs, the array achieves 90 % classification accuracy on a held-out test set. The array exhibits low sneak-path interference, attributed to high device nonlinearity in the OFF state and uniform switching thresholds across the matrix.

5. Discussion

5.1 Ionic Liquid-Mediated Switching Mechanism

The confined BMIM·PF_6 within the geopolymer pores provides mobile cations and anions that readily form and dissolve conductive filaments under very low electric fields. The imidazolium cation migrates toward the cathode under positive bias, reducing local activation energy for filament nucleation, while PF_6^- assists in stabilizing the filament backbone. This dual-ion contribution explains the forming-free behavior and sub-0.35 V set/reset thresholds observed across all microsphere sizes.

5.2 Influence of Microscale Geometry

Scaling the active element to 50–200 µm spheres concentrates the electric field at the electrode–sphere interface, promoting uniform filament growth pathways. The spherical geometry also minimizes edge defects and stress concentrations, which contributes to the low cycle-to-cycle variability (<10 %) and high

on/off ratios (>10^4). Moreover, soft-lithographic replica molding ensures tight control over element size and surface quality, critical for reproducible device performance.

5.3 Synaptic Function Realism and Energy Efficiency

Emulated short-term plasticity (PPF/PPD) and long-term plasticity (STDP) behaviors closely mirror biological synapses in both temporal dynamics and magnitude of conductance changes. The picojoule-scale energy per event (<3 pJ) places these GP–IL devices among the most energy-efficient synthetic synapses, making them highly attractive for large-scale spiking neural networks in edge-AI scenarios where energy budgets are stringent.

5.4 Array Integration and Sneak-Path Mitigation

Implementation of the 4×4 crossbar array demonstrates that GP–IL microsystems can be assembled into dense architectures without exotic selectors. The high nonlinearity of the OFF state and tight voltage threshold distributions minimize sneak-path currents, enabling accurate, on-device learning and pattern recognition. This suggests that larger arrays, when coupled with simple peripheral circuitry, can directly implement learning rules in hardware without additional selector devices.

5.5 Prospects for Flexible and Wearable Systems

The ambient-temperature synthesis and soft-lithographic processing are inherently compatible with flexible substrates and roll-to-roll manufacturing. Encapsulation in thin elastomer layers can preserve IL content while conferring mechanical resilience, opening the door to wearable neuromorphic sensors and bio-interfacing devices that process data locally and adapt in real time.

6. Conclusion

We have demonstrated a sustainable, microscale memristive platform by integrating geopolymer resonators with BMIM·PF_6 through a room-temperature, soft-lithographic process. Key achievements include:

- **Forming-free, low-voltage switching** at ±0.35 V with on/off ratios >10^4.
- **Biomimetic synaptic functions** (PPF, PPD, STDP) executed with <3 pJ per event.
- **Array-level learning** in a 4×4 crossbar achieving 90 % digit-recognition accuracy without additional selectors.

These findings position IL-functionalized geopolymer microsystems as a versatile, energy-efficient foundation for future neuromorphic hardware in edge and wearable applications. Future work will explore scaling to larger arrays, integration with flexible substrates, and co-design of learning algorithms to fully leverage the unique properties of this platform.

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