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Green Fabrication of Ionic Liquid-Enhanced Geopolymer Synapses via Solvent-Free Curing

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ABSTRACT

We report a sustainable, solvent-free route to fabricate ionic liquid–enhanced geopolymer synapses for low-power neuromorphic applications. Metakaolin-based geopolymer precursors were dry-mixed with 1-butyl-3-methylimidazolium tetrafluoroborate (BMIM·BF_4) at 10 wt % and cured at ambient conditions without added solvents or high-temperature firing. SEM and EDX mapping confirm homogeneous IL confinement within the geopolymer's interconnected pore network. Thermal analysis reveals stable IL retention up to 120 °C, while FTIR spectra verify chemical integrity post-curing. Au/geopolymer–IL/Au crossbar devices exhibit forming-free bipolar resistive switching with set/reset voltages of +0.50 V/–0.45 V and on/off ratios exceeding 10^2. Key synaptic behaviors—paired-pulse facilitation/depression and spike-timing-dependent plasticity—are emulated with energy consumption below 8 pJ per event. Devices maintain >10^3 switching cycles and >10^4 s retention without performance loss. Life-cycle assessment indicates a >60 % reduction in processing energy and solvent emissions compared to conventional solvent-based composites. This green fabrication strategy paves the way for scalable, eco-friendly neuromorphic hardware based on geopolymer–ionic liquid composites.

1. Introduction

The increasing demand for energy-efficient computing has driven the exploration of alternative memory technologies that integrate storage and processing. Traditional silicon-based memristors often rely on high-temperature deposition and toxic solvents, which undermine environmental and economic sustainability. Geopolymers—aluminosilicate materials synthesized under ambient conditions from industrial byproducts—offer a low-carbon, low-energy scaffold with tunable porosity and mechanical robustness, making them attractive hosts for next-generation memory devices.

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Ionic liquids (ILs) are non-volatile molten salts with high ionic conductivity and wide electrochemical windows. When incorporated into porous matrices, ILs facilitate ion migration and redox reactions under reduced electric fields, resulting in forming-free switching at sub-volt thresholds. Embedding ILs within geopolymers combines the environmental benefits of ambient curing with the electrochemical versatility of ILs, potentially enabling sustainable, low-power synaptic elements.

Conventional methods for integrating ILs into solid hosts often employ solvents to promote infiltration, followed by solvent evaporation or high-temperature annealing. These approaches increase energy consumption, release volatile organic compounds, and complicate scale-up. A solvent-free, dry-mixing and curing protocol eliminates these drawbacks, reducing processing steps, emissions, and energy use while preserving IL functionality and device performance.

This study introduces a green fabrication strategy for ionic liquid–enhanced geopolymer synapses via solvent-free curing. Our key contributions are:

1. **Eco-friendly processing:** Development of a dry-mixing and ambient-cure method to integrate BMIM·BF_4 into geopolymer matrices without solvents or high-temperature steps.
2. **Device characterization:** Demonstration of forming-free bipolar resistive switching at ±0.5 V with on/off ratios >10^2 in Au/geopolymer–IL/Au crossbar devices.
3. **Neuromorphic functionality:** Emulation of paired-pulse facilitation/depression and spike-timing-dependent plasticity with energy per event <8 pJ.
4. **Life-cycle benefits:** Quantification of processing energy and emissions savings (>60 %) relative to solvent-based composite fabrication.

By combining sustainable materials and green processing, our approach lays the groundwork for scalable, eco-friendly neuromorphic hardware based on geopolymer–ionic liquid composites.

2. Literature Review

Efforts to develop sustainable, high-performance memory devices have increasingly turned toward composite materials that marry robust inorganic scaffolds with mobile ionic species. [1-2] Geopolymers—aluminosilicate networks formed under ambient conditions from industrial byproducts—offer a particularly attractive platform, combining low-energy synthesis, tunable porosity, and excellent mechanical stability.[3] Their open, interconnected pore structures can be tailored to host functional additives without the need for high-temperature sintering or energy-intensive processing, addressing key environmental concerns associated with conventional ceramic or polymeric hosts. However, achieving uniform incorporation of active species into these porous matrices remains a challenge, especially when the target is a sub-micron or microscale memory element.[4-5]

Ionic liquids (ILs) have emerged as versatile dopants for resistive switching applications, owing to their high ionic conductivity, negligible vapor pressure, and wide electrochemical stability windows. When confined within a solid matrix, ILs can facilitate filamentary conduction through reversible redox reactions and ion migration under reduced electric fields, enabling forming-free operation and sub-volt switching thresholds. Past approaches to integrate ILs into porous hosts have typically relied on solvent-based infiltration or thermal annealing to promote penetration and remove excess solvent, but these methods incur significant energy costs and generate volatile emissions. Moreover, the risk of IL phase separation or chemical degradation during high-temperature steps can undermine device reliability and lifetime, limiting the practical translation of these materials into scalable neuromorphic hardware.[6-8]

Solvent-free processing strategies—centered on dry mixing of solid IL precursors with host powders followed by ambient curing—offer a promising route to circumvent these drawbacks. By eliminating liquid solvents and high-temperature steps, dry mixing preserves IL chemical integrity while simplifying

manufacturing and reducing carbon emissions. Ambient-cure geopolymer composites formed via such methods retain the host's pore architecture and achieve homogeneous IL distribution, as evidenced by uniform elemental mapping and consistent electrical response across multiple devices. Crucially, these green-fabricated composites exhibit stable IL confinement, with minimal volatilization or loss under prolonged thermal or electrical stress.[9] The resulting devices demonstrate reliable bipolar resistive switching with low set/reset voltages, high on/off ratios, and emulation of synaptic behaviors such as paired-pulse facilitation, depression, and spike-timing-dependent plasticity at picojoule energy scales.[10-11]

Despite these advances, fully solvent-free, room-temperature approaches to create geopolymer–IL synapses have been underexplored in the context of device integration and array-level functionality. Scaling from single-element prototypes to crossbar architectures introduces further complexity, including alignment precision, sneak-path mitigation, and learning-rule implementation. Addressing these issues within a green fabrication framework could unlock a new class of eco-friendly, energy-efficient neuromorphic systems, with applications ranging from edge-AI accelerators to wearable bio-interfaces.[12]

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 Solvent-Free Mixing and Curing Protocol

1. **Dry mixing:** Metakaolin powder and sodium silicate solution (pre-mixed into a paste) were combined in a 1:1.5 mass ratio and homogenized in a planetary ball mill at 150 rpm for 2 h. $BMIM \cdot BF_4$ ionic liquid was then added directly to the dry geopolymer powder at 10 wt % relative to the total solid mass and mixed by gentle tumbling for 30 min to achieve uniform coating.
2. **Pellet pressing:** The dry composite powder was loaded into a 10 mm diameter stainless-steel die and uniaxially pressed at 200 MPa for 5 min to form 0.5 mm-thick pellets.
3. **Ambient curing:** Pressed pellets were transferred to a humidity-controlled chamber (relative humidity ≈ 80 %) and cured at room temperature (22 °C) for 48 h. No solvent evaporation or thermal firing steps were used. After curing, pellets were demolded and stored in a desiccator until use.

3.3 Device Fabrication

1. **Substrate preparation:** Glass slides were cleaned sequentially in acetone, isopropanol, and deionized water, then dried under nitrogen.
2. **Bottom electrode deposition:** A 5 nm Cr adhesion layer and 100 nm Au were thermally evaporated through a shadow mask to form parallel lines (width = 100 μm, spacing = 100 μm).

3. **Composite integration:** Geopolymer–IL pellets were gently placed atop the bottom electrode array and pressed under 1 MPa for 1 min to ensure intimate contact.
4. **Top electrode deposition:** Orthogonal Au lines (100 nm) were evaporated through a second mask, creating crossbar junctions with active areas of 100 µm × 100 µm.
5. **Encapsulation:** A thin polydimethylsiloxane (PDMS) layer (~10 µm) was spin-coated over the entire device to inhibit IL volatilization during testing.

3.4 Characterization Methods

- **Morphology & composition:**
  - SEM/EDX: Cross-sectional and surface imaging to assess pore structure and verify BMIM·BF_4 distribution (mapping of B and F signals).
- **Chemical analysis:**
  - FTIR: Spectra collected from 400 to 4000 cm^{-1} to detect imidazolium and BF_4^- functional groups.
  - TGA: Mass loss measured from 25 °C to 250 °C at 10 °C min^{-1} under N_2 to quantify IL retention.
- **Electrical measurements:**
  - **I–V Sweeps:** Bipolar voltage sweeps (–0.6 V to +0.6 V) at 0.01 V s^{-1} using a semiconductor parameter analyzer to extract set/reset voltages and on/off ratios.
  - **Pulse Testing:** Synaptic protocols (±0.5 V, 10 ms pulses) for paired-pulse facilitation/depression and spike-timing-dependent plasticity measurements.
  - **Endurance and Retention:** Endurance cycling (10^3 set/reset pulses at ±0.5 V, 100 µs width) and retention monitoring at 0.1 V read bias for 10^4 s.
- **Life-Cycle Assessment:**
  - **Energy Consumption:** Measurement of processing energy for mixing, pressing, and curing compared to a benchmark solvent-based protocol.
  - **Emission Estimates:** Calculation of solvent vapor and CO_2-equivalent emissions avoided by eliminating solvents and high-temperature steps.

4. Results

4.1 Morphology and Composition

SEM imaging of the green-fabricated geopolymer–IL pellets reveals a dense network of angular aluminosilicate grains interconnected by a continuous pore structure. Pore diameters range from 50 to 200 nm, consistent across the pellet cross-section. EDX elemental maps show uniform distributions of boron and fluorine signals throughout the geopolymer matrix, confirming homogeneous confinement of BMIM·BF_4 within the pore network rather than surface aggregation. No micron-scale IL pools or phase separations are observed, indicating that the dry-mixing and ambient-cure protocol effectively embeds IL into the host without solvent assistance.

4.2 Chemical Integrity and Thermal Stability

FTIR spectra of cured geopolymer–IL composites retain the characteristic imidazolium ring vibrations at 1570 and 1450 cm^{-1}, as well as BF_4^- stretching modes near 1050 cm^{-1}, demonstrating that BMIM·BF_4

remains chemically intact post-curing. TGA analysis shows a single mass-loss event between 110 °C and 180 °C, corresponding to IL volatilization. The measured IL content of 9.6 ± 0.4 wt % (vs. nominal 10 wt %) confirms minimal loss during curing. After one thermal cycle to 100 °C, IL loss is under 2 wt %, indicating strong retention within the geopolymer network and suitability for low-temperature electronic operation.

4.3 Resistive Switching Performance

Au/geopolymer–IL/Au crossbar devices exhibit forming-free bipolar resistive switching under low-voltage sweeps. Representative I–V curves (±0.6 V sweep at 0.01 V s^{-1}) show a set transition at +0.50 V and a reset transition at –0.45 V. Read currents measured at 0.1 V are ~2 × 10^{-7} A (ON state) and ~1 × 10^{-9} A (OFF state), yielding an on/off ratio of ~200. Across 30 devices, the average set voltage is +0.52 ± 0.06 V and the reset voltage is –0.47 ± 0.05 V, with device-to-device variability below 12 %.

4.4 Synaptic Functionality

Under paired-pulse protocols (±0.5 V, 10 ms pulses), the second EPSC peak is 1.20× the first (50 ms interval), indicating paired-pulse facilitation (PPF index = 20 %). With reversed polarity pulses, paired-pulse depression (PPD index = 25 %) is observed. Spike-timing-dependent plasticity (STDP) measurements, using ±0.5 V, 10 ms pulses with delays ranging from –50 ms to +50 ms, produce asymmetric conductance changes: potentiation up to +15 % for pre-before-post delays and depression up to –12 % for post-before-pre delays. Energy consumption per synaptic event, computed as $E = V \cdot I \cdot t$, remains below 8 pJ for both potentiation and depression pulses.

4.5 Endurance and Retention

Endurance cycling over 1,000 set/reset pulses (±0.5 V, 100 µs width) demonstrates stable ON and OFF currents with drift under 5 %. Retention tests at a 0.1 V read bias show that both conductance states are maintained for more than 10,000 s, with ON-state decay less than 10 % and OFF-state current remaining below 1 × 10^{-9} A throughout.

4.6 Life-Cycle Assessment

Processing energy for the solvent-free protocol (dry milling, pressing, 48 h ambient-cure) was measured at 3.2 kWh kg^{-1} of composite, compared to 8.5 kWh kg^{-1} for a benchmark solvent-based infiltration route (including solvent drying and low-temperature annealing). Estimated solvent-related emissions are eliminated entirely in the green route, while CO_2-equivalent emissions are reduced by 65 % per kilogram of composite produced. These metrics underscore the environmental benefit of the solvent-free process without compromising device performance.

5. Discussion

The solvent-free dry-mixing and ambient-cure approach successfully embeds ionic liquid within the geopolymer host, achieving uniform distribution and strong retention without resorting to solvents or high-temperature steps. The confined BMIM·BF_4 facilitates mobile-ion–mediated resistive switching, lowering activation barriers so that forming-free bipolar transitions occur at ±0.5 V. The on/off ratio of ~200 and tight threshold distributions (±0.06 V) reflect consistent filament dynamics across devices, attributable to the homogeneous IL coating and preserved pore architecture.

Synaptic behaviors closely mimic biological short- and long-term plasticity: paired-pulse facilitation/depression indices (20–25 %) and STDP asymmetry (±12–15 %) align with neural models, while picojoule energy per event (<8 pJ) meets the demands of energy-constrained edge-AI systems. Durability metrics—1,000 cycles and $>10^4$ s retention—highlight that the geopolymer network effectively traps IL, preventing volatilization or chemical degradation during repeated electrical stress.

From an environmental perspective, the green fabrication route reduces energy consumption by over 60 % and eliminates solvent emissions, positioning geopolymer–IL synapses as a sustainable alternative to conventional memristive composites. The ambient-condition curing simplifies manufacturing and enables seamless integration with flexible substrates or roll-to-roll processes, broadening application potential to wearable neuromorphic sensors and adaptive interfaces.

Comparison to solvent-based protocols underscores the trade-off between processing complexity and device performance: while higher IL loadings or annealing can marginally improve on/off ratios, they incur greater energy and emissions costs. Our optimized 10 wt % IL loading and room-temperature curing strike a practical balance, delivering robust neuromorphic functionality with minimal environmental impact.

6. Conclusion

We have demonstrated a fully solvent-free, green fabrication method for ionic liquid–enhanced geopolymer synapses. Key outcomes include:

1. **Eco-friendly processing:** Dry mixing and ambient curing embed BMIM·BF_4 at 10 wt % into metakaolin geopolymers without solvents or firing.
2. **Low-voltage switching:** Forming-free bipolar resistive transitions occur at ±0.50 V/–0.45 V with on/off ratios $>10^2$.
3. **Biomimetic plasticity:** PPF, PPD, and STDP behaviors are emulated with <8 pJ per event, and devices endure $>10^3$ cycles with $>10^4$ s retention.
4. **Environmental benefits:** Processing energy is reduced by 65 % and solvent emissions are eliminated compared to benchmark solvent-based composites.

This work establishes geopolymer–ionic liquid composites as a sustainable platform for scalable neuromorphic hardware. Future studies will explore array integration on flexible substrates, dynamic learning-rule implementations, and extension to biocompatible ILs for medical-grade sensing applications.

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