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Room-Temperature Ionic Liquid-Doped Ceramic Composites for Low-Voltage Synaptic Devices

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ABSTRACT

Room-temperature processing of ionic liquid–doped ceramic composites offers a sustainable, low-cost route to neuromorphic devices that operate at sub-volt switching thresholds. In this work, we report the fabrication and characterization of Al_2O_3–SiO_2 ceramic membranes functionalized with 1-ethyl-3-methylimidazolium triflate ($EMIM^+·OTf^-$) via a simple, solvent-free mixing and curing protocol at ambient temperature. Morphological analysis (SEM, EDX) confirms uniform IL dispersion within the ceramic pore network, while thermogravimetric and contact-angle measurements demonstrate enhanced moisture retention and surface wettability. Electrical testing of Au/EMIM-ceramic/Au cross-bar devices reveals bipolar resistive switching with set and reset voltages as low as +0.45 V and –0.40 V, respectively, and an on/off ratio exceeding 10^3. Synaptic functionalities—including paired-pulse facilitation, paired-pulse depression, and spike-timing-dependent plasticity—are emulated with energy consumption per event below 5 pJ. Devices retain stable conductance states for over 10^4 s and endure more than 10^3 switching cycles without performance degradation. Compared to undoped ceramics, EMIM doping reduces activation barriers and stabilizes conductive filament formation under low electric fields. These findings highlight the promise of room-temperature IL–ceramic composites for scalable, energy-efficient in-memory computing and pave the way for integrated neuromorphic architectures in flexible and wearable electronics.

1. Introduction

Memristive devices—often dubbed the "fourth fundamental circuit element"—have attracted intense interest for their ability to emulate synaptic behavior in hardware and enable in-memory computing.

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Traditional oxide-based memristors (e.g., TiO_2, HfO_2) typically require high-temperature deposition and forming steps, which add cost and limit integration onto flexible substrates. Recent work has demonstrated that porous ceramics and geopolymers can serve as low-cost, sustainable hosts for resistive switching when functionalized with mobile ions or dopants . Ionic liquids (ILs), with their negligible vapor pressure and high ionic conductivity, are promising candidates for room-temperature doping to control filament formation and charge transport in these matrices.

While IL-functionalized geopolymer memristors have shown sub-volt operation and long retention, their ceramic analogues remain underexplored. Ceramics such as Al_2O_3–SiO_2 offer superior mechanical stability and high thermal endurance compared to geopolymers, yet their dense microstructure can impede ion mobility unless engineered with appropriate pore networks. Furthermore, most reports on IL/ceramic composites rely on solvent-based infiltration or high-temperature annealing, which compromises IL integrity. A simple, solvent-free, room-temperature route to uniformly load IL into ceramic hosts would enable true low-cost fabrication of synaptic devices.

In this work, we address these challenges by developing a one-step, ambient-temperature mixing and curing process to incorporate 1-ethyl-3-methylimidazolium triflate (EMIM·OTf) into Al_2O_3–SiO_2 ceramic membranes. Our specific contributions are:

1. Materials innovation: Demonstration of solvent-free IL loading into a ceramic pore network at room temperature.
2. Device performance: Achievement of bipolar resistive switching at sub-0.5 V thresholds, with on/off ratios $>10^3$.
3. Synaptic emulation: Implementation of key neuromorphic functions (PPF, PPD, STDP) with energy per event <5 pJ.
4. Durability: Evidence of $>10^3$ switching cycles and $>10^4$ s retention without degradation.

Together, these results establish IL-doped ceramic composites as a compelling platform for scalable, low-voltage synaptic devices in next-generation neuromorphic systems.

2. Literature Review

Memristive switching in ceramic matrices has been widely investigated as an alternative to conventional metal-oxide devices. Porous and composite ceramics provide tunable pathways for ion migration, enabling reversible formation and rupture of conductive filaments under applied bias. These structures often demonstrate high on/off ratios and nonlinear I–V characteristics, making them suitable for nonvolatile memory and logic-in-memory applications.[1-2] However, dense ceramic hosts typically require high-temperature treatments to achieve the desired defect concentrations or phase compositions, which limits compatibility with flexible or polymeric substrates.[3-4]

Ionic liquids (ILs) have emerged as versatile dopants and functional agents to enhance electrochemical activity in solid matrices. Their high ionic conductivity, negligible vapor pressure, and broad electrochemical windows facilitate low-voltage charge transport when confined within porous hosts. In polymer and geopolymer systems, IL incorporation has been shown to lower activation barriers for ion migration, improve moisture retention, and stabilize switching behavior over extended periods. Translating these benefits to ceramic systems demands careful control over pore architecture and IL distribution to prevent phase separation or IL loss during processing and device operation.[5-8]

Room-temperature, solvent-free methods for IL loading into solid hosts promise a streamlined fabrication pathway for synaptic devices. Techniques based on dry mixing, mechanical compaction, or mild curing can preserve IL integrity while achieving homogeneous distribution within the ceramic network.[9] Such approaches eliminate the need for high-temperature annealing or lengthy solvent evaporation steps, reducing energy consumption and simplifying scale-up.[10] When combined with tailored pore-formers or templating agents, these methods can yield composites with interconnected channels for facile ion transport and robust mechanical stability.[11-12]

Collectively, these advances suggest that ceramic–IL composites fabricated via ambient-temperature protocols could unite the mechanical and thermal advantages of ceramics with the electrochemical versatility of ILs.[13-14] By leveraging simple, scalable processing, it becomes possible to realize low-voltage synaptic devices that are both durable and energy-efficient, addressing key challenges in neuromorphic hardware development.[15-16]

3. Experimental Section

3.1 Materials

Alumina (Al_2O_3, average particle size ~0.5 µm) and fumed silica (SiO_2, specific surface area ~200 $m^2\ g^{-1}$) powders were procured and used as the ceramic host. The ionic liquid 1-ethyl-3-methylimidazolium triflate (EMIM·OTf) was selected for its high ionic conductivity and negligible vapor pressure. Gold (Au) pellets (99.99 %) were used to form top and bottom electrodes. All reagents were used as received without further purification.

3.2 Composite Preparation

Ceramic powders were dried at 120 °C for 12 h to remove residual moisture, then mixed in a 70:30 wt % Al_2O_3:SiO_2 ratio. EMIM·OTf was added at 15 wt % relative to total ceramic mass. The mixture was dry-milled in a planetary ball mill at 200 rpm for 4 h using zirconia balls, ensuring uniform IL coating on ceramic particles. The resulting powder was pressed into 0.5 mm-thick membranes (Ø 10 mm) under 100 MPa uniaxial pressure and cured at room temperature for 24 h to allow IL redistribution within the pore network.

3.3 Device Fabrication

Cross-bar devices were assembled by depositing 100 nm Au bottom electrodes onto cleaned glass substrates via thermal evaporation through a shadow mask (line width = 100 µm). The IL–ceramic membrane was laminated onto the bottom electrode under slight pressure (~1 MPa). Top electrodes (100 nm Au) were then deposited orthogonally, forming active junction areas of 100 µm × 100 µm.

3.4 Characterization Methods

- **Morphology and Composition:** Scanning electron microscopy (SEM) coupled with energy-dispersive X-ray spectroscopy (EDX) was used to image cross-sectional pore structure and verify IL distribution.
- **Thermogravimetric Analysis (TGA):** Samples (≈10 mg) were heated from 25 to 300 °C at 10 °C min^{-1} under N_2 to quantify IL content and retention.
- **Contact-Angle Measurements:** Static water contact angles were recorded to assess surface wettability before and after IL doping.
- **Electrical Testing:** I–V sweeps and pulse measurements were performed on a semiconductor parameter analyzer. Bipolar switching was characterized using voltage sweeps from –1 V to +1 V

at 0.01 V s^{-1}. Retention tests monitored device current at read voltage (0.1 V) over 10^4 s. Endurance cycling was conducted with ±0.5 V pulses (100 µs width) up to 10^3 cycles.
- **Synaptic Emulation:** Paired-pulse facilitation and depression were measured by applying paired voltage pulses (±0.5 V, 10 ms width) with variable inter-pulse intervals. Spike-timing-dependent plasticity was evaluated by pre- and post-synaptic pulse trains (±0.5 V, 10 ms) with controlled delay times.

4. Results

4.1 Morphology and Composition

SEM cross-sections of the IL–ceramic membranes reveal an interconnected pore network with pore diameters ranging from 100 to 300 nm. The ceramic grains are uniformly coated by a thin IL film, as evidenced by EDX elemental maps showing a homogeneous distribution of fluorine (from OTf^-) and nitrogen (from $EMIM^+$) throughout the pore walls. No macroscopic cracks or phase separations are observed, indicating that the 15 wt % IL loading and room-temperature curing preserve membrane integrity.

4.2 Ionic-Liquid Retention and Wettability

TGA traces show a single mass loss step between 100 °C and 200 °C corresponding to IL volatilization, with an IL content of 14.7 ± 0.3 wt %—in close agreement with the nominal loading. After thermal cycling to 100 °C and back, IL loss is under 2 %, demonstrating strong retention within the pore network. Contact-angle measurements decrease from 75° ± 2° (undoped ceramic) to 28° ± 3° (IL–ceramic), confirming enhanced surface wettability and suggesting improved ion accessibility at the electrode interfaces.

4.3 Resistive Switching Characteristics

Au/IL-ceramic/Au cross-bar devices exhibit bipolar switching with forming-free behavior. Typical I–V sweeps (–1 V to +1 V) show a set transition at +0.45 V (ON state) and a reset at –0.40 V (OFF state), with a low read voltage of 0.1 V. The ON/OFF current ratio exceeds 10^3 at 0.1 V. Statistical analysis across 20 devices yields average set/reset voltages of +0.47 ± 0.05 V and –0.42 ± 0.04 V, respectively, with device-to-device variability under 12 %.

4.4 Synaptic Emulation

Paired-Pulse Facilitation (PPF): When two consecutive +0.5 V, 10 ms pulses are applied with a 50 ms inter-pulse interval, the second peak current is 1.32 × the first, indicating short-term memory behavior. The PPF index decays exponentially with increasing interval, consistent with biological synapses.

Paired-Pulse Depression (PPD): Under reversed polarity (–0.5 V pulses), the second response is suppressed to 0.78 × the first, demonstrating bidirectional plasticity.

Spike-Timing-Dependent Plasticity (STDP): Pre- and post-synaptic pulse trains (±0.5 V, 10 ms width) separated by delays from –50 ms to +50 ms produce asymmetric weight updates. Positive delays (pre before post) yield potentiation up to +22 % conductance change, while negative delays induce depression down to –15 %.

4.5 Energy Consumption and Durability

Energy per synaptic event, calculated from the product of applied voltage, read current, and pulse duration, is below 5 pJ for both potentiation and depression pulses. Endurance testing over 10^3 set/reset cycles at ±0.5 V (100 µs pulse width) shows negligible drift in ON/OFF currents (<5 %). Retention measurements at 0.1 V read bias confirm stable conductance states for over 10^4 s with less than 8 % state decay.

5. Discussion

5.1 Role of Ionic-Liquid Doping

Incorporation of EMIM·OTf at 15 wt % into the ceramic host dramatically lowers the activation barrier for filament formation. The IL's mobile ions and improved moisture retention facilitate easier redox reactions and ionic migration pathways under low electric fields, accounting for the sub-0.5 V set/reset thresholds observed. The thin IL films lining the pore walls act as ion reservoirs, stabilizing filament growth and rupture during switching, which explains the forming-free behavior and low cycle-to-cycle variability.

5.2 Comparison to Undoped Ceramic

Undoped Al_2O_3–SiO_2 membranes require >2 V forming steps and exhibit high variability in switching voltages due to limited intrinsic defect concentrations. In contrast, IL-doped composites show uniform, reproducible switching at ambient conditions. The enhanced surface wettability lowers contact resistance at the electrode interface, further reducing the effective voltage needed to trigger resistive transitions.

5.3 Neuromorphic Implications

Emulation of short-term plasticity (PPF, PPD) and long-term plasticity (STDP) under low-energy pulses indicates that IL–ceramic synapses can mimic key biological functions with picojoule-scale energy budgets. The asymmetric potentiation/depression window and exponential decay characteristics closely follow neural models, making these devices attractive for hardware implementations of spiking neural networks.

5.4 Durability and Integration Potential

Robust retention over 10^4 s and endurance beyond 10^3 cycles demonstrate that IL confinement within the pore network prevents rapid IL loss or decomposition under repeated electrical stress. The room-temperature, solvent-free process is compatible with flexible substrates and roll-to-roll manufacturing, paving the way for large-area arrays. Future work on crossbar integration and interface engineering could further enhance device density and uniformity.

6. Conclusion

This study establishes a facile, room-temperature route to fabricate ionic liquid–doped Al_2O_3–SiO_2 ceramic composites for low-voltage synaptic devices. Key achievements include:

- Low-voltage switching at ±0.5 V with on/off ratios $>10^3$ and forming-free operation.
- Energy-efficient synaptic functions (PPF, PPD, STDP) realized with <5 pJ per event.
- Durable performance, evidenced by $>10^3$ switching cycles and $>10^4$ s retention without significant drift.

The synergy of mechanical robustness, thermal stability, and electrochemical versatility positions IL–ceramic composites as a promising platform for scalable, energy-efficient neuromorphic hardware. Future efforts will focus on crossbar array demonstrations, dynamic learning algorithm implementation, and exploration of bio-compatible ceramic–IL systems for wearable applications.

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