Machine Learning-Assisted High-Throughput Exploration of Lead-Free Perovskite Compositions for Memristor Devices

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1. Introduction

The demand for efficient and scalable computing architectures has surged as digital transformation accelerates. Traditional von Neumann computing faces performance bottlenecks due to the separation of memory and processing units, leading to inefficiencies in data transfer. Memristors, which integrate memory and processing functionalities, have emerged as a promising solution to enable in-memory computing and neuromorphic applications.[1]

Among various materials investigated for memristors, halide perovskites have demonstrated great potential due to their highly tunable optoelectronic properties, facile processability, and especially unique ionic migration characteristics.[2]While lead-based halide perovskites were initially explored for memristive applications, concerns over toxicity have driven interest in lead-free alternatives.[3] However, the range of halide perovskite materials reported for memristors remains limited, and there is no comprehensive study exploring wider material space for device applications.

Here, we establish a high-throughput experimental platform and protocol capable of screening a vast array of halide perovskite compositions for memristor and neuromorphic applications directly in device configurations. This approach enables systematic exploration and direct comparison of the performance landscape, paving the way for discovering new material candidates with optimized properties for next-generation memory and computing technologies.

2. Experiments and results overview

To systematically investigate the vast material space for memristor applications, we have developed a high-throughput experimental platform that integrates device fabrication with screen-printing and automated precursor mixing, comprehensive electrical characterization, and machine learning-driven optimization (Fig. 1).

2.1 Device Fabrication and Material Screening

Our high-throughput device fabrication process utilizes a screen-printing technique to pattern an array of 81 scaffold cells on fluorine-doped tin oxide (FTO) glass substrates. Each cell consists of a mesoporous ZrO_2 layer serving as a hollow spacer and a carbon layer as an electrode. The active layer is formed by infiltrating the scaffold cells with precursor solutions, followed by controlled crystallization to yield diverse halide-based memristor materials.

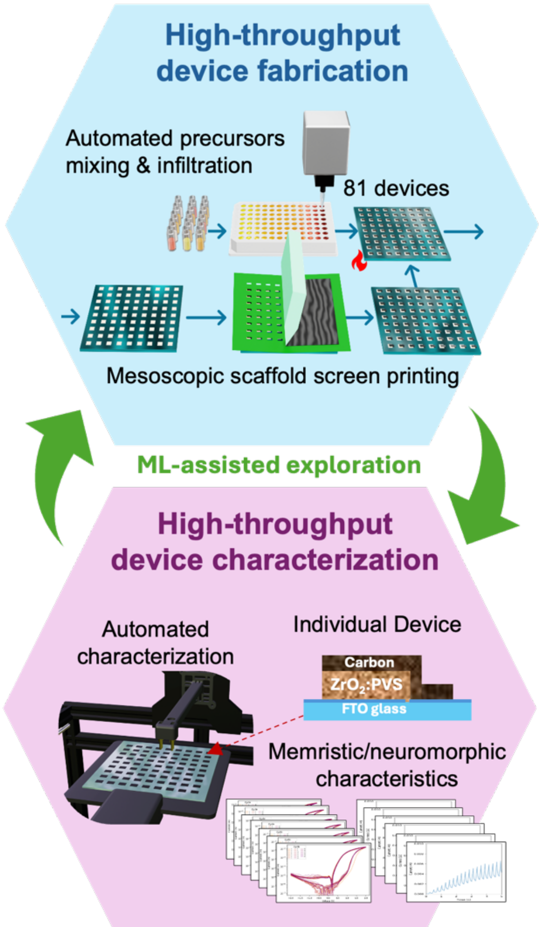

Fig. 1: Experimental verification of AI results is a big plus, but is not mandatory

To explore a broad composition space, we employed a liquid handling robot for precise mixing of 19 precursor solutions, encompassing organic halide salts (MAI, MABr, MACl, FAI, FABr, AVAI, GI, BAI, PEAI) and metal halides (AgI, AgBr, CuI, CuBr, CsI, CsBr, RbCl, BiI_3, $BiBr_3$, SbI_3). The initial composition screening was designed using Latin Hypercube Sampling (LHS), ensuring broad coverage of material space. With minimal compositional constraints, it enables the formation of a diverse range of end-product materials, including 2D and 3D perovskites, perovskite-inspired structures (e.g., pnictohalides), and materials with stoichiometric variations (excess/deficient atomic configurations).

2.2 Memristor Characterization and Optimization

The electrical performance of the fabricated devices was characterized through multiple metrics, including current-voltage (I-V) hysteresis, short-term and long-term potentiation/depression, and

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impedance spectroscopy. To systematically refine material compositions and processing parameters, we implemented Bayesian Optimization to identify formulations that maximize linear potentiation and enhance the number of distinct conductivity states. This data-driven approach not only accelerates material discovery but also provides insights into the fundamental mechanisms governing superior neuromorphic behavior.

Our platform represents a significant advancement in high-throughput material exploration for memristors, facilitating the rapid identification of promising candidates for neuromorphic computing and next-generation memory applications.

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