Lead-free Bio-Inkjet Printing with Bulk KNN Actuators and AITL-based Adaptive Control

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Abstract—This paper proposes a biocompatible inkiet printing architecture based on lead-free piezoelectric actuators, specifically bulk KNN (K,Na)NbO₃, combined with chip-on-film (COF) driver ICs, silicon cavity integration, and an AITL (Adaptive Intelligent Three-Layer) supervisory control framework. Unlike conventional PZT-driven industrial heads, the KNN approach supports biocompatibility and Pb-free compliance, while moderate performance (3–5 pL droplets, 10^6 shots, ± 50 V operation) satisfies bioprinting needs. Beyond actuation, the electromechanical response of KNN is leveraged for self-diagnosis: detecting missing droplets and estimating ink viscosity. This diagnostic data feeds into an AITL architecture—an inner PID loop for real-time stability, an FSM layer for mode transitions and compensation, and an LLM-driven supervisory layer for adaptive redesign of control rules. We present the architecture, process flow, and representative biomedical applications, highlighting the potential of KNN as both actuator and sensor.

I. INTRODUCTION

Inkjet printing has become a versatile technology in both industrial and consumer applications, with piezoelectric lead zirconate titanate (PZT) actuators serving as the dominant driver mechanism. Despite extensive research on lead-free piezoelectrics such as (K,Na)NbO₃ (KNN) and Bi_{0.5}Na_{0.5}TiO₃ (BNT), these alternatives have not replaced PZT in mainstream printing due to insufficient compatibility in key metrics such as piezoelectric coefficient d_{33} , long-term durability, and costeffective manufacturing.

In biomedical applications, however, the design requirements differ fundamentally. Rather than extreme durability and maximum actuation strength, biocompatibility, chemical safety, and moderate performance are the primary needs. Typical bio-printing tasks—including cell patterning, protein array generation, and hydrogel dispensing—require droplet volumes in the range of 1–10 pL and endurance on the order of 10^6 shots, which are significantly less demanding than industrial printing standards.

This work therefore explores a Bio-Inkjet (Bio-IJ) architecture based on bulk KNN actuators, combined with chipon-film (COF) driver integration and silicon cavity bonding. By tailoring the system to biomedical rather than industrial requirements, the proposed approach provides a feasible lead-free alternative that aligns with environmental regulations and biological safety.

II. BACKGROUND: PB-FREE PIEZOELECTRICS

Research on lead-free piezoelectrics has been pursued for more than two decades as part of global efforts to replace lead zirconate titanate (PZT) with environmentally benign alternatives. Among the most prominent candidates are (K,Na)NbO₃ (KNN), Bi_{0.5}Na_{0.5}TiO₃ (BNT), and Sc-doped AlN (ScAlN).

KNN typically exhibits a piezoelectric coefficient d_{33} in the range of 150–250 pm/V, with optimized compositions reaching up to 300–400 pm/V, and a Curie temperature exceeding 400 °C. These properties make it suitable for moderate-displacement actuators. In contrast, ScAlN provides a more modest d_{33} of 20–30 pm/V, but its excellent CMOS compatibility and feasibility for thin-film MEMS integration have positioned it as an attractive material for micro-scale devices. BNT and related systems have also been investigated, though issues such as depolarization and processing complexity have limited their adoption.

Despite these advances, industrial printing applications have not embraced Pb-free piezoelectrics. The primary obstacle has been the expectation of *full PZT compatibility*—requiring identical performance in terms of strain output, durability over 10^9 cycles, and manufacturing cost. Such stringent requirements exceed the current capabilities of KNN, BNT, or ScAlN.

In addition to performance considerations, PZT carries an intrinsic **toxicity risk** due to its high lead content. Although industrial printheads encapsulate PZT to prevent contact with ink, *crack formation or delamination could expose Pb to the fluidic environment*. This risk is particularly unacceptable for biomedical printing, where contamination by lead would compromise cell viability, protein function, and regulatory compliance. Consequently, Pb-free piezoelectrics are not only an environmental imperative but also a **safety requirement** for Bio-IJ applications.

III. PROPOSED ARCHITECTURE

The proposed Bio-Inkjet (Bio-IJ) actuator system is designed to balance biocompatibility, manufacturability, and sufficient actuation performance for biomedical applications. Its main components are as follows:

 Bulk KNN multilayer stack actuator: A 200–500 μm thick piezoelectric stack providing moderate displacement suitable for picoliter-scale droplet ejection.

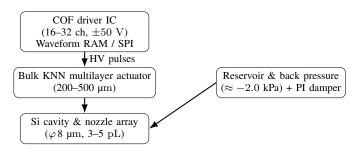


Fig. 1. System architecture of the proposed Bio-Inkjet (Bio-IJ). A bulk KNN actuator, COF high-voltage driver, silicon cavity/nozzles, and fluidics (back-pressure with PI damper) are integrated.

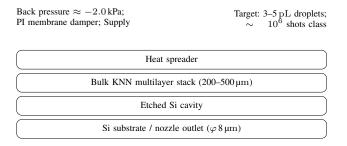


Fig. 2. Cross-sectional schematic of the Bio-IJ printhead. The KNN stack is bonded to a silicon cavity with φ 8 μ m nozzles; fluidics provide back-pressure stabilization and damping.

- COF driver IC: A chip-on-film high-voltage driver with 16–32 channels, operating at up to ±50 V, and incorporating waveform RAM with SPI control for flexible pulse shaping.
- Si cavity and nozzle array: A silicon-etched cavity directly bonded to the actuator, with nozzles of $\varphi 8 \, \mu m$ diameter producing droplets in the 3–5 pL range.
- Reservoir and back pressure control: A fluidic supply stabilized at approximately −2.0 kPa using a PID-controlled regulator to ensure consistent meniscus positioning.
- PI membrane damper: A polyimide-based damping layer integrated in the reservoir to suppress pressure fluctuations and prevent satellite droplets.

The associated *process flow* begins with fabrication of the bulk KNN stack and electrode finishing, followed by terminal cutting and COF assembly. The actuator is then mounted with a heat spreader for thermal management and finally bonded to the silicon cavity structure, resulting in an integrated Bio-IJ printhead module.

A. Self-Diagnosis and AITL Control

The electromechanical response of the bulk KNN actuator can be exploited for self-diagnosis and adaptive control. Surrogate signals derived from drive current and charge increments correlate with droplet volume and viscosity, enabling online monitoring.

Two representative strategies are highlighted:

- Self-diagnosis: Detecting missing droplets by charge/displacement thresholds, estimating viscosity from relaxation dynamics.
- Adaptive control: Updating PID gains and FSM transitions in real time based on surrogate measurements, while keeping safety constraints (drive voltage ≤ ±50 V).

Detailed mathematical models, including recursive least squares (RLS) identification, MIT-rule adaptation, and viscosity mapping, are provided in the Appendix.

IV. APPLICATIONS

The proposed Bio-Inkjet architecture enables several key biomedical applications in which moderate actuation performance, precise droplet control, and biocompatibility are prioritized over extreme durability:

- Cell patterning: Controlled deposition of living cells into predefined patterns for tissue engineering and regenerative medicine. Gentle actuation and droplet volumes of 1– 10 pL support high cell viability, with survival rates above 80% reported under comparable shear stress conditions.
- **Protein and DNA microarrays**: Picoliter-scale dispensing of biomolecules onto functionalized substrates for high-throughput screening, diagnostics, and drug discovery. The ability to generate uniform, sub-100 μm spots is critical for assay reproducibility.
- Hydrogel 3D printing: Layer-by-layer deposition of biocompatible hydrogels, followed by UV or thermal curing, to fabricate soft scaffolds for cell culture and organ-onchip platforms. Precise droplet placement ensures structural fidelity and material homogeneity.

These use cases demonstrate that the moderate performance of bulk KNN actuators—picoliter droplet generation at voltages below ± 50 V—is sufficient to meet biomedical requirements. Here, droplet volume control, biocompatibility, and integration with fluidic handling systems are far more critical than the billion-cycle endurance demanded in industrial printing.

V. DROPLET FORMATION SIMULATION

To provide an educational yet illustrative understanding of droplet ejection, a simplified capillary–inertial nozzle model was implemented. The model uses lumped parameters (nozzle radius, viscosity, surface tension, density, and trapezoidal drive pulses) to approximate meniscus velocity and ejected volume.

Representative time traces at 10, 30, and 50 V show the meniscus velocity profiles (Fig. 4).

VI. MATERIAL PERFORMANCE COMPARISON

Table I compares the four piezoelectric material systems considered for inkjet actuation.

While PZT offers the highest actuation, the presence of lead precludes its use in direct biomedical environments. KNN achieves moderate performance with Pb-free compliance, making it a promising candidate for Bio-IJ systems.

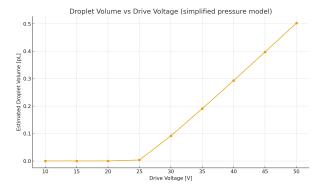


Fig. 3. Estimated droplet volume versus drive voltage using the simplified lumped model (5-50 V).

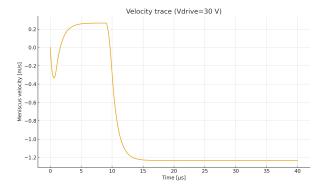


Fig. 4. Example meniscus velocity trace for 30 V drive pulse.

VII. BIO-APPLICATION SUITABILITY MAP

Suitability scores (1–5) were heuristically assigned to each material for representative biomedical applications (Fig. 5).

VIII. CONCLUSION

This paper has proposed a Bio-Inkjet (Bio-IJ) architecture based on bulk KNN actuators as a lead-free alternative to conventional PZT-based printheads. By combining multilayer KNN stacks, COF driver ICs, and silicon cavity integration, the system achieves picoliter-scale droplet generation under moderate voltages while ensuring material biocompatibility.

Unlike industrial printing, where full PZT compatibility in terms of maximum d_{33} , billion-cycle endurance, and cost efficiency is required, biomedical printing places emphasis on safety, controlled droplet volume, and operational reliability over shorter lifetimes. The proposed approach aligns well with these requirements, providing sufficient performance for

 $\label{table I} \textbf{TABLE I} \\ \textbf{INDICATIVE PERFORMANCE COMPARISON OF PIEZOELECTRIC MATERIALS.} \\$

Material	d ₃₃ (pm/V)	T_c (°C)	Bio Safety
PZT	300-600	250-350	Pb risk
KNN	150-300	>400	Pb-free
BNT	100-200	200-300	Pb-free
ScAlN	20–30	>600	Pb-free

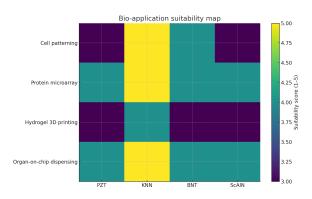


Fig. 5. Suitability map (score 1-5) of PZT, KNN, BNT, and ScAlN for various Bio-IJ applications.

applications such as cell patterning, protein microarrays, and hydrogel 3D fabrication.

These findings highlight bulk KNN as a practical foundation for standardizing lead-free Bio-IJ systems in research, clinical, and educational domains. Future work will involve experimental validation of droplet formation, long-term reliability testing under bio-relevant conditions, and system integration with existing bioprinting workflows.

Most importantly, this work emphasizes that in biomedical inkjet printing, safety and biocompatibility must take precedence over extreme performance metrics, positioning KNN-based Bio-IJ as a safe and viable path toward Pbfree bioprinting.

APPENDIX A AITL CONTROL DERIVATIONS

A. Surrogate Sensing and Plant Model

During a drive pulse u_k , the KNN stack current i(t) and charge $q(t) = \int i(t)dt$ define surrogate features:

$$y_k \triangleq \alpha_1 q_k^{\text{peak}} + \alpha_2 \Delta q_k + \alpha_3 t_{r,k}, \tag{1}$$

with calibration coefficients α_i .

B. Inner-Loop Dynamics

We adopt a discrete 2nd-order model:

$$y_{k+1} = a_1(\eta)y_k + a_2(\eta)y_{k-1} + b_0(\eta)u_k + b_1(\eta)u_{k-1} + d_k,$$
 (2)

where viscosity η influences $\{a_i, b_i\}$.

C. Recursive Least Squares (RLS)

The parameter update follows:

$$\theta_k = \theta_{k-1} + K_k(y_k - \phi_k^{\mathsf{T}} \theta_{k-1}),$$
 (3)

$$K_k = \frac{P_{k-1}\phi_k}{\lambda + \phi_k^{\top} P_{k-1}\phi_k},\tag{4}$$

$$P_k = \lambda^{-1} (P_{k-1} - K_k \phi_k^{\top} P_{k-1}), \tag{5}$$

with forgetting factor $\lambda \in (0, 1]$.

D. Adaptive PID Examples

IMC-PID Retuning:

$$K_p = \frac{\tau + \theta}{K(\lambda + \theta)}, \quad K_i = \frac{K_p}{\tau + \theta}, \quad K_d = \frac{\tau \theta}{2(\tau + \theta)}.$$
 (6)

MIT Rule:

$$\theta_{k+1} = \theta_k - \Gamma \psi_k e_k, \quad \psi_k = \frac{\partial \hat{y}_k}{\partial \theta}.$$
 (7)

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AUTHOR BIOGRAPHY

Shinichi Samizo received the M.S. degree in Electrical and Electronic Engineering from Shinshu University, Japan. He worked at Seiko Epson Corporation as an engineer in semiconductor memory and mixed-signal device development, and contributed to inkjet MEMS actuators and PrecisionCore printhead technology. He is currently an independent semiconductor researcher focusing on process/device education, memory architecture, and AI system integration.