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Integrated Organic Pyroelectric Sensors

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Abstract

Organic pyroelectric sensors employing poly (vinylidene fluoride–trifluoroethylene) [P(VDF-TrFE)] were fabricated and comprehensively characterized to realize a mechanically flexible and electronically integrable infrared-detection platform. Through a γ-butyrolactone – based sol–gel synthesis followed by controlled thermal annealing, highly crystalline β-phase copolymer films were achieved on PET substrates. Ferroelectric hysteresis characterization exhibited a coercive field of approximately 80 MV m^{-1} and a saturation polarization close to 200 MV m^{-1}, indicative of pronounced dipole alignment and stable remanent polarization. Optimization of the poling conditions – specifically electric fields near 150 MV m^{-1} and temperatures around 130 °C – resulted in significantly enhanced pyroelectric voltage and current outputs. Frequency-domain analysis demonstrated excellent correspondence with theoretical RC-network predictions, delineating distinct cut-off behaviors for current- and voltage-mode operation. Furthermore, the incorporation of a graphite absorber layer, in conjunction with square-wave laser modulation, markedly increased thermal-to-electrical conversion sensitivity. Collectively, these findings substantiate the feasibility of low-temperature, solution-processable fabrication routes for organic pyroelectric devices, providing exceptional mechanical compliance and seamless integration with organic thin-film transistor (OTFT) readout circuitry. This platform establishes a promising route toward scalable, large-area, and conformable infrared-sensing architectures suited for next-generation flexible electronic systems.

Keywords: copolymer, flexible, pyroelectric, polarization, ferroelectric.

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İnteqrasiya edilmiş üzvi yarımkeçirici piroelektrik sensorlar

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Xülasə

Poli (viniliden flüorid–trifloroetilen) [P(VDF-TrFE)] əsaslı üzvi piroelektrik sensorlar elastik, inteqrasiyaolunan və infraqırmızı şüaları yüksək həssaslıqla təyin edən platformanın formalaşdırılması məqsədilə hazırlanmış və çoxşaxəli funksional təhlildən keçirilmişdir. γ-butirolakton əsasında həyata keçirilən sol-gel sintezi və mərhələli termal tablama prosesi nəticəsində PET altlıqlar üzərində yüksək dərəcədə kristal quruluşa və β-fazalı morfologiyaya malik kopolimer təbəqələr sintez edilmişdir. Ferroelektrik histerezis analizləri təqribən 80 MV/m-də koersiv sahənin və 200 MV/m-də isə maksimal polyarizasiyasının dəyərlərini göstərmiş, materialın daxilində dipol momentlərinin yüksək istiqamətlənmə dərəcəsini və onun güclü sahə təsirlərində stabil vəziyyətdə qalan polyarizasiyasını göstərmişdir. Poling prosesinin optimallaşdırılması – xüsusilə ~150 MV/m elektrik sahəsi və ~130 °C temperatur rejimi altında – piroelektrik gərginlik və cərəyan siqnallarının amplitudunu əhəmiyyətli şəkildə artırmış, yükün yığılma kinetikasının və dipol re-oriyentasiyasının effektivliyini yüksəltmişdir. Tezlik xarakteristikasının analizi nəzəri RC-ekvivalent sxem modelləri ilə yüksək uyğunluq göstərmiş, həm cərəyan-rejim, həm də gərginlik-rejim iş prinsipləri üçün müxtəlif kəsilmə tezliklərinin qiymətinin dəyişmə aralığını aydın şəkildə göstərmişdir. Qrafit əsaslı uducu təbəqənin inteqrasiyası və düzbucaqlı-dalğa formalı lazer modulyasiyasının tətbiqi nəticəsində istilik-elektrik çevrilmə effektivliyi və piroelektrik həssaslıq göstəriciləri nəzərəçarpacaq dərəcədə yüksəlmişdir. Əldə edilən nəticələr aşağı temperaturlu, məhlul-əsaslı emal texnologiyalarının üzvi piroelektrik elementlərin geniş sahəli və mexaniki elastik istehsalında tam tətbiq oluna biləcəyini empirik şəkildə təsdiqləyir. Bu yanaşma yüksək mexaniki elastiklik, geniş səthlərə inteqrasiyası və üzvi nazik təbəqəli tranzistor (ÜST) əsaslı mikrosxem əsaslı arxitekturaları ilə tam uyğunluq göstərərək, gələcək nəsil yüksək elastikli və adaptiv infraqırmızı sensor sistemlərinin yaradılması üçün elmi və texnoloji əsas formalaşdırır.

Açar sözlər: kopolimer, elastik, piroelektrik, polyarizasiya, ferroelektrik.

Интегрированные органические полупроводниковые пироэлектрические сенсоры

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Аннотация

Органические пироэлектрические сенсоры на основе сополимера поли(винилиденфторида–трифторэтилена) [P(VDF–TrFE)] были изготовлены и всесторонне охарактеризованы с целью создания гибкой и интегрируемой платформы инфракрасного детектирования. Применение сол-гель метода с использованием γ-бутиролактона и последующего контролируемого термического отжига позволило сформировать высококристаллические β-фазные плёнки сополимера на подложках из ПЭТ. Ферроэлектрические петли гистерезиса продемонстрировали коэрцитивное поле порядка 80 МВ/м и насыщенную поляризацию около 200 МВ/м, что свидетельствует о выраженной ориентации дипольных моментов и высокой степени доменной стабильности. Оптимизация условий поляризации — при электрических полях около 150 МВ/м и температурах порядка 130 °C — обеспечила значительное усиление пироэлектрических откликов по току и напряжению, а также повышение эффективности перераспределения зарядов и переориентации диполей. Частотный анализ диэлектрического отклика показал высокое соответствие теоретическим моделям RC-эквивалентных цепей, чётко выявив характерные граничные частоты для токового и вольт-режимов работы. Введение графитового абсорбирующего слоя совместно с модуляцией лазерного излучения в форме прямоугольных импульсов позволило существенно повысить чувствительность термоэлектрического преобразования. Совокупность полученных результатов подтверждает технологическую осуществимость низкотемпературных, растворных процессов формирования органических пироэлектрических устройств. Такие структуры обладают высокой механической гибкостью, совместимы с органическими тонкоплёночными транзисторными схемами считывания и открывают перспективы масштабируемых, конформных инфракрасных сенсорных систем нового поколения.

Ключевые слова: кополимер, эластичный, пироэлектрический, поляризация ферроэлектрический

Introduction

Infrared (IR) sensors enable the detection of thermal radiation across the mid- to far-IR spectrum. Of particular relevance is the ~10 μm band, where the black-body emission of living organisms' peaks at room temperature, making this window exceptionally valuable for presence, motion, and bio-related sensing. Broadly, IR detection technologies fall into two families that differ in mechanism, bandwidth, and operating constraints.

The first family comprises photon (quantum) detectors, which are intrinsically wavelength-selective. Depending on device physics and readout scheme, they leverage the photovoltaic, photoconductive, or photoelectric effects. These detectors are commonly formed from narrow-bandgap semiconductors – such as indium antimonide (InSb) – and are renowned for their very high sensitivity and ultrafast response. Their impressive performance, however, comes with an energetic threshold: they require incident photons above a material-specific minimum energy. As a result, to suppress thermally generated carriers and meet stringent noise targets, photon detectors are frequently operated at cryogenic temperatures. The associated cooling burden adds cost and complexity, especially for large-area, portable, or conformable systems.

The second family consists of thermal detectors, which infer incident radiation indirectly by registering a temperature rise in the sensing element. This temperature change manifests as a variation in electrical properties – most commonly a resistance change (bolometers) or a thermoelectric voltage (thermocouples and thermopiles). Thermal detectors respond more slowly than quantum detectors, yielding lower-frequency bandwidths, but offer a key advantage: because their signal depends on absorbed heat rather than on discrete photon energies, they are broadly sensitive across the IR spectrum. In practice, their usable spectral range is limited chiefly by the transmittance of the entrance window and optical train, not by an electronic bandgap. The trade-off is that, in absolute sensitivity, thermal detectors are generally inferior to photon detectors by several orders of magnitude. Within this class, pyroelectric detectors occupy a special position as the fastest thermal detectors, since the underlying signal originates directly from temperature-induced polarization changes at the molecular scale.

Pyroelectricity denotes the generation of an electrical response in a material upon a change in temperature. It arises in dielectrics that possess a spontaneous or "frozen-in" polarization due to oriented dipole moments. Such behavior is found in ten crystalline point groups, as well as in certain ceramics and in polymers that have undergone appropriate processing to align dipoles or induce ferroelectric phases. The phenomenon has been recognized since antiquity - Theophrastus offered what is likely the earliest description in On Stones—but polymer pyroelectricity, though identified in the 1940s, remained weak and of limited practical interest for decades. A pivotal advance came in 1971, when Bergmann and co-workers reported strong pyroelectricity in poly (vinylidene fluoride) (PVDF), thrusting polymers to the forefront of pyroelectric research and applications. Rapidly thereafter, Glass and colleagues and Yamaka demonstrated polymer-based pyroelectric IR sensors, and Bergmann and Crane introduced a pyroelectricity-enabled xerography process. Since then, the physics of pyroelectricity in polymers has been substantially clarified comprehensively reviewed by Bauer and Lang – and significant pyroelectric responses have been documented in a wide variety of amorphous, semicrystalline, single-crystalline, and liquid-crystalline polymer systems.

A complete pyroelectric polymer pixel is typically conceived as a capacitive pyroelectric stack interfaced to optical and electronic subsystems [1]. The input unit encompasses IR absorption and focusing – often via engineered absorption layers and micro-optical elements – to concentrate radiant power into the active film. The output unit implements signal readout, impedance transformation, and amplification tailored to the high source impedance and charge-based signals of pyroelectric elements [2]. Commercial designs illustrate this architecture's versatility. For example, Siemens' angle-selective motion sensor (marketed as PID-21) employs PVDF as a freestanding array mounted on a supporting frame and hybrid-connected to its electronics on a printed-circuit board [3]. An alternative path is monolithic or hybrid integration on silicon, wherein polymer pyroelectric arrays are fabricated directly over CMOS readout circuits that provide impedance matching and gain. While attractive for compactness and noise control, such silicon-centric integration confronts difficult thermal-management constraints: the high thermal conductivity and heat-sinking capacity of silicon can diminish temperature rise in the sensing film, lowering responsivity unless careful thermal isolation strategies are employed [3,4].

These integration constraints have historically steered "classical" integrated IR detector systems toward rigid, planar substrates – a sensible compromise for many imaging tasks but one that precludes continuous panoramic (360°) views and limits deployment on curved or compliant surfaces [4,5]. Against this backdrop, organic thin-film transistors (OTFTs) emerge as enabling companions for polymer pyroelectric sensors. Both device classes can be processed at low temperatures over large areas on flexible substrates (e.g., polymer foils), using cost-effective manufacturing routes compatible with roll-to-roll approaches. This shared processability invites co-fabrication of the sensing element and its front-end electronics on the same flexible platform, yielding integrated, conformable pyroelectric sensor systems. Such systems could unlock application domains difficult for rigid silicon to address: pedestrian protection and vulnerable-road-user detection in automotive safety; human–machine interfaces in mobile and wearable electronics; large-area security and occupancy monitoring; low-cost home electronics; and electronic skin concepts for robotics and prosthetics.

Despite this promise, and as earlier reports have emphasized, the translation to large-area, fully integrated organic pyroelectric sensor modules have been gated by demanding OTFT performance requirements. Readout transistors must combine low-voltage operation, extremely high input impedance, minimal leakage, and long-term stability – characteristics that have only recently approached the thresholds needed for robust, manufacturable systems. Consequently, while polymer pyroelectric sensing and OTFT electronics are each well-established individually, their seamless integration at scale remains a challenging but fertile frontier. We therefore target the outstanding gap – scalable, flexible integration of polymer pyroelectric pixels with high-impedance, low-leakage OTFT readout – by demonstrating a PVDF–OTFT platform with engineered thermal isolation for large-area IR sensing.

Structure of Poly (Vinylidene Fluoride)

Poly (vinylidene fluoride) (PVDF) exhibits a suite of functional properties that make it attractive for diverse applications. Depending on processing history, PVDF crystallizes into several polymorphs – commonly denoted α, β, γ, and δ – whose chain conformations govern polarity. Vinylidene fluoride monomers carry a sizable molecular dipole moment ($\mu_v \approx 7 \times 10^{-30}\ C \cdot m$) arising from charge separation between the electropositive hydrogens and electronegative fluorines in the –CF_2–CH_2– repeat unit. In the ferroelectric β-phase, the chains adopt the all-trans (TTTT) conformation, aligning the local dipoles along a single polar axis; because the crystalline packing orients this polar axis coherently across chains, the material exhibits a macroscopic spontaneous polarization on the order of 130 $\frac{\mu C}{m^2}$ [6]. Because ferro-, piezo-, and pyroelectric responses require a robust macroscopic spontaneous polarization, maximizing the fraction of the ferroelectric β-phase is a central processing objective. Whether PVDF solidifies into the non-polar (paraelectric) α-phase or the polar (ferroelectric) β-phase depends sensitively on thin-film fabrication conditions. A practical route to obtain the β-phase directly from the melt is to introduce the copolymer poly (trifluoroethylene) (PTrFE), yielding P(VDF-TrFE). Substituting a hydrogen with the larger fluorine in the TrFE comonomer stabilizes the all-trans conformation, thereby favoring β-phase formation. From a molecular-level standpoint, the copolymer's individual repeat unit is less polar than that of PVDF, which in principle lowers the theoretical upper bound of spontaneous polarization per unit volume. In practice, however, P(VDF-TrFE) often displays higher effective ferroelectric performance because the copolymer increases overall crystallinity and stabilizes the polar phase fraction in semicrystalline films.

Formation of P(VDF–TrFE) Active Layers and Baseline Capacitance Structures

Prior studies showed that freeze-dried (lyophilized) PVDF gels prepared with γ-butyrolactone (GBL) promote the TT chain conformation and thereby the β-phase. Motivated by this, we translated the GBL-based sol–gel route from PVDF to P (VDF-TrFE) as a low-temperature spin-on strategy to yield highly crystalline, sufficiently flat ferroelectric thin films [7,8].

In the process, GBL is heated to 180 °C under reflux to suppress solvent loss. P (VDF-TrFE) pellets with VDF:TrFE ratios of 55:45, 65:35, and 76:24 are introduced and dissolved, and the solution is held ~2 h at temperature. After cooling to room temperature, the solution is transferred to a glass bottle to gel. Immediately before coating, the viscous gel is gently warmed to re-form a low-viscosity sol for spinning. By tuning spin speed and solid content, film thicknesses of ~300 nm to 3 µm are readily achieved [7,8]. Typical dielectric constants are 15–17 for 55:45 compositions and 10–12 for 65:35 and 76:24 materials.

For electrical evaluation of ferroelectric behavior, we fabricated capacitance-type test structures using standard microfabrication steps. Aluminum (50 nm) was sputtered onto glass or PET (Melinex) substrates to form the bottom electrode, followed by spin-coating of the P(VDF-TrFE) sol. The films then underwent a 110 °C calcination/anneal for 5 h, which enhanced crystallinity, and were slow-cooled to room temperature. Top electrodes of silver (50–80 nm) were thermally evaporated through shadow masks. Polarization–electric-field hysteresis and remanent polarization were subsequently measured in a Sawyer–Tower configuration.

Ferroelectric Characteristics of P (VDF-TrFE)

In Fig. 1 at left, a typical hysteresis loop measurement of a P (VDFTrFE) capacitor is highlighted.

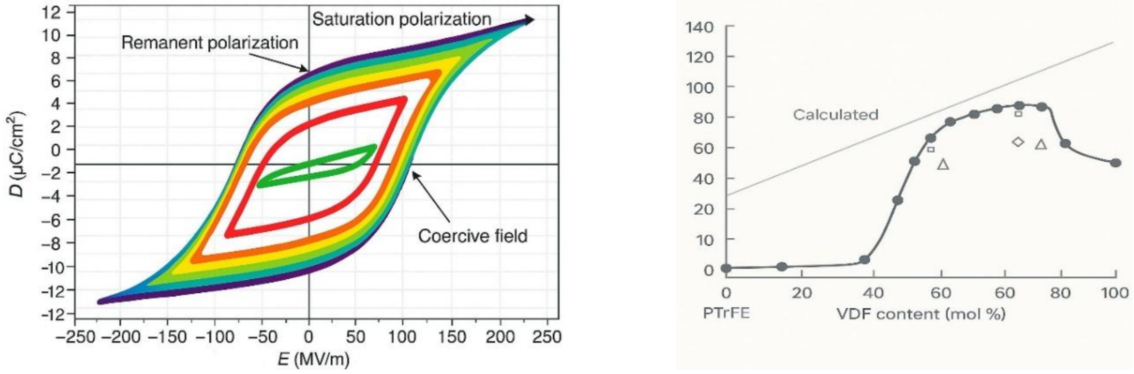
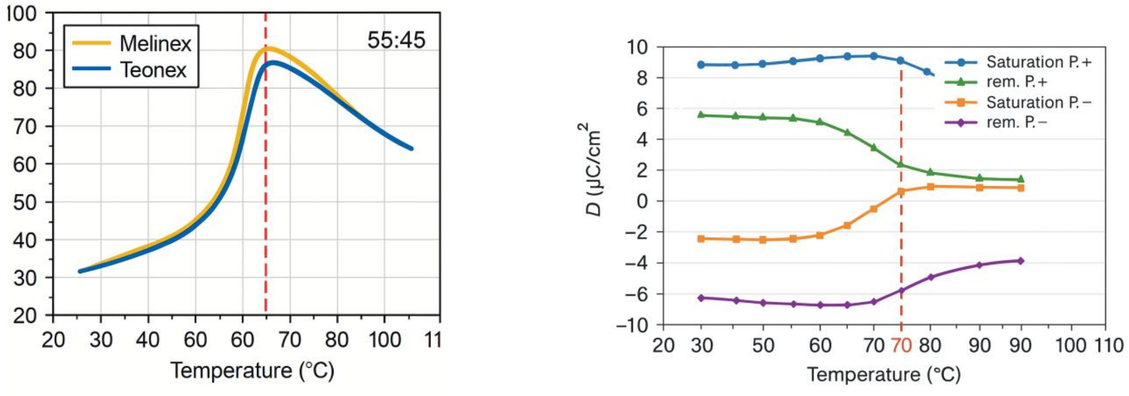


Figure 1 – Typical hysteresis loop of a flexible P(VDF-TrFE) sensor element:(right) and remanent polarization (left)

Hysteresis loops recorded with progressively higher maximum fields show a coercive field of roughly 80 MV/m and a saturation polarization near 200 MV/m. In Fig. 1 (right), theory and experiment (per Furukawa) map the remnant polarization Pr versus PVDF fraction, revealing the largest P_r at 50–80% PVDF. At very low PVDF, too few polar units suppress polarization and ferroelectricity; at very high PVDF, the α-phase is favored and crystallinity falls below ~50%, both lowering polarizations. Between 80% and 50% PVDF, P_r declines gradually because the average dipole moment drops (a TrFE unit carries about half the dipole of a VDF unit). Sol-gel P(VDF-TrFE) copolymers align with the highest (Furukawa) P_r values reported, whereas many other studies yield smaller values, indicating weaker ferroelectric behavior.

All ferroelectrics exhibit a Curie temperature (T_c) marking the transition to the paraelectric phase. Even if chains remain in the all-trans conformation, macroscopic spontaneous polarization vanishes at Tc due to loss of chain alignment. This appears in the polarization–temperature curves where Pr → 0 above T_c (Fig. 2b). The temperature dependence of the permittivity likewise shows a clear transition at T_c; for 55% VDF, heating and cooling traces overlap, consistent with a second-order transition (Fig. 2a).



(a) (b)

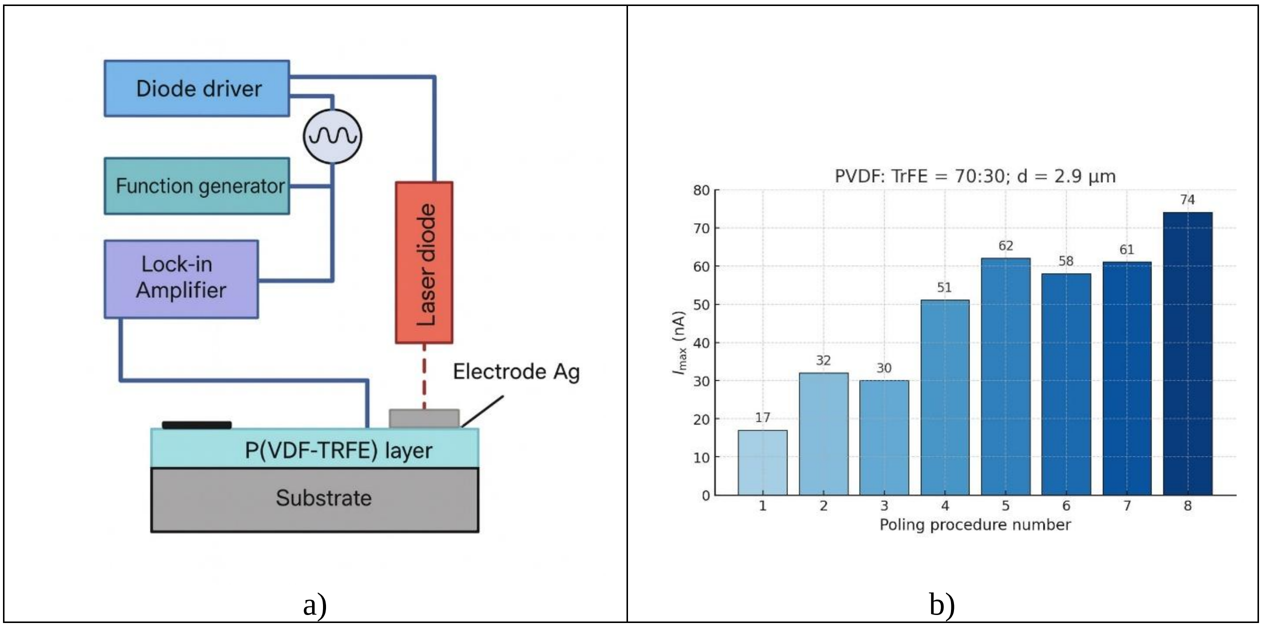
Figure 2 – (a) Dielectric constant versus temperature for a flexible P(VDF-TrFE) (55:45) sensor element; the dashed line marks the phase-transition temperature during cooling and heating. (b) For the same flexible P(VDF-TrFE) element, dielectric constant versus temperature, with the transition temperature identified at the point where the remanent polarization goes to zero.

Instrumentation and Measurement Framework for Pyroelectric Response Evaluation

To accurately assess the pyroelectric effect, the dipoles within the material must be oriented in a uniform direction to induce a measurable macroscopic spontaneous polarization. This alignment process is referred to as poling. Poling is accomplished by applying an external electric field across the sample via the electrodes, with the applied field exceeding the material’s coercive field threshold.

For P(VDF–TrFE) films, the coercive field is approximately 80 MV/m, so the poling field must be substantially higher – typically around 150 MV/m - to ensure complete dipole alignment. A stepwise voltage escalation method is often employed, incorporating intermediate zero-voltage intervals to stabilize the polarization process, as reported in previous studies.

The effectiveness of poling is governed not only by the magnitude of the applied electric field but also by the temperature during the poling procedure. As illustrated in Fig. 3a for a 2.9 µm-thick film containing 70 % VDF, optimal polarization – and consequently the highest pyroelectric current and voltage response - is achieved when the field is gradually increased to about 140 MV/m while maintaining the sample temperature near 130 °C.



a) b)

Figure 3 – a) Setup for the measurement of the pyroelectric response. b) İnfluence of poling voltage, poling temperature, laser excitation waveform and absorber structure on the maximum of the pyroelectric current and voltage response

Dynamic Behavior of the Pyroelectric Response

The pyroelectric response, recorded using the experimental configuration illustrated in Figure 3a, can be evaluated in both voltage and current detection modes. The frequency-dependent characteristics of these signals for a copolymer film with a thickness of 2.9 µm are presented in Figure 3b. Under the employed lock-in amplifier configuration, the maximum voltage response occurs at approximately 5 Hz, whereas the maximum current response is observed within the frequency range of 2–6 kHz. This behavior can be explained using the equivalent circuit representation and the relation

$$|V_{pyro}| = |I_{pyro}| \cdot \frac{R}{\sqrt{1+(\omega RC)^2}},$$

$$C = C_i + C_p,$$

$$R = \frac{R_p R_i}{R_p + R_i}$$

Which simplifies to the expression given in eq. (1). Accordingly, the voltage response scales proportionally with IR at frequencies below the cut-off ($\omega < \omega_c$) and as I/(ωC) at higher frequencies ($\omega > \omega_c$). The cut-off frequency ($\omega_c = 1/RC$) is dictated by the overall RC time constant of the circuit, where R_p and C_p represent the intrinsic resistance and capacitance of the pyroelectric element, and R_i and C_i denote the input resistance and capacitance of the measurement setup, respectively.

Inspection of figure 3 reveals that, in addition to the dipole alignment parameters (poling voltage and temperature), both the optical absorption characteristics of the material (presence of an absorber layer) and the excitation waveform substantially affect the magnitude of the pyroelectric response. Incorporating a graphite absorber layer more than doubles the voltage output, while substituting a square-wave laser modulation enhances the response by roughly 30 % under identical poling conditions (e.g., 500 V).

For quantitative assessment, the voltage and current sensitivities of the pyroelectric device are obtained by normalizing the measured response to the incident radiant power. As demonstrated in figure 4 for a thin pyroelectric sensor fabricated on a Melinex substrate, the expected inverse frequency relationship between voltage and current responses (for $\omega > \omega_c$) is clearly observed consistent with the theoretical prediction from eq.

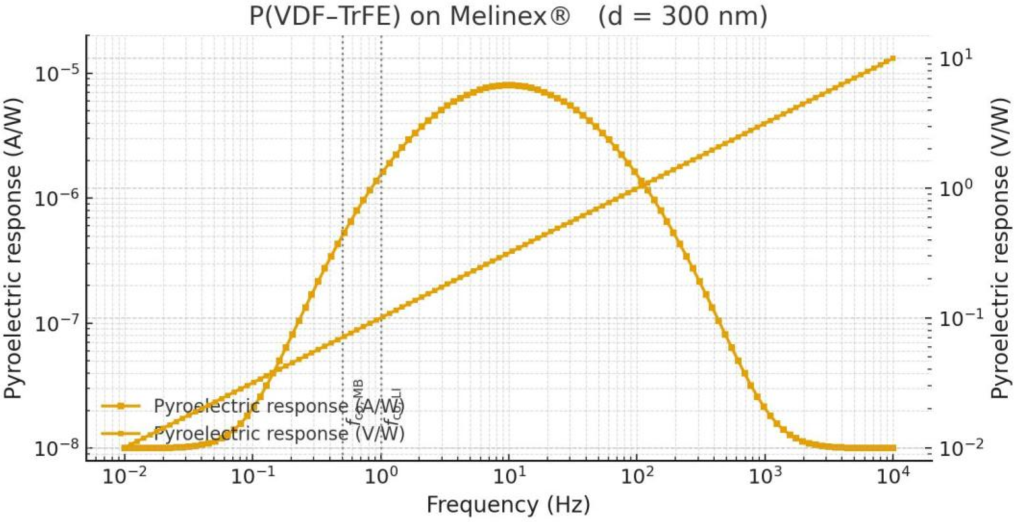


Figure 4 – Pyroelectric voltage and current behavior of 300 nm VDF:TrFE (55:45) layers deposited on PET (Melinex) substrate.

Conclusion

This study demonstrates the successful fabrication and functional evaluation of flexible organic pyroelectric sensors based on P(VDF–TrFE) copolymer thin films prepared via a γ-butyrolactone sol–gel process. The resulting β-phase-rich structures exhibited strong ferroelectric and pyroelectric responses, as evidenced by well-defined hysteresis loops, high remanent polarization, and frequency-dependent voltage–current behavior consistent with theoretical RC modeling. The integration of optimized poling conditions and absorber structures significantly improved the sensor’s thermal-to-electrical transduction efficiency, confirming the potential of P(VDF–TrFE) films for reliable infrared detection on flexible substrates.

Beyond validating a scalable low-temperature processing route, this work highlights the compatibility of organic pyroelectric films with organic thin-film transistors (OTFTs), opening the path toward fully integrated, large-area, and conformable infrared sensing systems. Such architectures are particularly promising for wearable electronics, aerospace monitoring, robotic tactile interfaces, and intelligent environmental sensing.

Future investigations also should prioritize:

1) Advanced hybrid integration of pyroelectric layers with high-impedance OTFT readouts,
2) Nanostructured absorbers for spectral selectivity and higher responsivity,

3) Multi-functional sensor networks combining pyroelectric, piezoelectric, and thermoelectric modalities, and
4) AI-assisted modeling and system optimization for real-time adaptive detection.

Altogether, these directions define a clear trajectory toward next-generation flexible infrared platforms capable of merging sensitivity, mechanical compliance, and intelligent signal processing.

Conflict of Interests

The author declares there is no conflict of interests related to the publication of this article.

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