

Synthesis, Characterization, and Applications of Schiff Base Flexible Organic Crystals

Kurdo Abdulkhaleq Abdullah¹, Karzan Mahmood Ahmed² , Aryan Fathulla Qader^{1,*} ,
Rebaz Anwar Omer^{1,3} , Eman Ibraheem Abdulkareem¹ 

1 Department of Chemistry, Faculty of Science and Health, Koya University, Danielle Mitterrand Boulevard, Koya KOY45, Kurdistan Region–F.R., Iraq; kurdoabdullah22@gmail.com (K.A.A.); aryan.qader@koyauniversity.org (A.F.Q.); rebaz.anwar@koyauniversity.org (R.A.O.); eman.ibraheem@koyauniversity.org (E.I.A.);

2 Department of Chemistry, College of Education, University of Garmian, 46021 Kalar, Kurdistan Region – F.R. Iraq; karzan.mahmood@garmian.edu.krd;

3 Department of Pharmacy, College of Pharmacy, Knowledge University, Erbil 44001, Iraq; rebaz.anwar@koyauniversity.org;

* Correspondence: aryan.qader@koyauniversity.org;

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Abstract: The advent of flexible organic crystals has transformed the way we view molecular crystals, which were previously considered brittle. This has sparked significant interest in exploring organic crystalline materials that are mechanically adaptable for advanced purposes. Schiff base compounds, known for their straightforward synthesis, high yield, responsiveness to stimuli, and strong mechanical attributes, emerge as top contenders in this domain. This comprehensive review covers recent progress in flexible organic crystals derived from Schiff bases, beginning with their accidental discovery and progressing to complex design techniques and a broad range of applications. Some applications include biomimetic soft robotics, organic photonic integrated circuits, biomimetic soft robots, optical waveguides, and optoelectronic devices. Notably, the investigation of multifunctional applications at the microscopic level has been made possible by the employment of atomic force microscopy-micromanipulation methods. Understanding container patterns of different Schiff base flexible organic crystals driven by non-covalent interactions and their direct association with mechanical characteristics is the main goal of this molecular-level crystal engineering investigation into mechanical flexibility. This knowledge synthesis intends to guide the creation of novel flexible organic crystals that combine distinctive properties and encourage continued investigation of mechanically adaptable organic crystalline materials for various technological and biomedical applications.

Keywords: flexible organic crystals; Schiff base compounds; mechanical properties; crystal engineering; non-covalent interactions.

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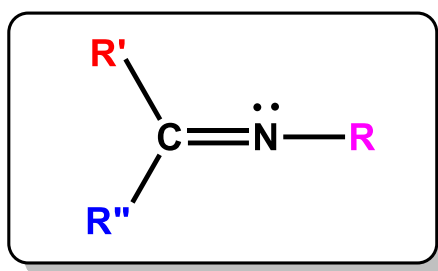
1. Introduction

The conventional perception of molecular crystals as inflexible and delicate structures endured until the advent of flexible organic crystals, which contradicted this notion [1]. This discovery triggered a sudden increase in curiosity and motivated further investigation into flexible organic crystalline materials, marking the beginning of a new era for intelligent materials. In the last ten years, extensive research has been done to create innovative materials that have enhanced mechanical properties and versatile capacities [2]. Schiff base compounds

are leading prospects for flexibility because of their ease of synthesis, high yield, reactivity to external stimuli, and outstanding mechanical characteristics. This paper discusses the recent progress on flexible organic crystals with Schiff bases that are elastic, plastic, and hybrid. The article discusses the path of Schiff-based flexible organic crystals from accidental discovery to state-of-the-art design, highlighting its many applications in many fields [3, 4]. Mechanical flexibility is a critical physical property that significantly affects material performance and is essential in practical applications. Traditionally, elastomers, thin polymer films, or liquid crystals have been relied upon for this purpose [5]. Organic single crystals have traditionally been seen as hard and fragile structures due to their highly organized and regular pattern, dense arrangement, and inflexible overall shape [6, 7] despite their particular advantages in the realm of optoelectronics, such as reduced defects, homogenous composition, lightweight design, high electron transport capacity, and enhanced carrier mobility. Recently, bendable organic crystals have been discovered, challenging the conventional understanding of molecular crystals [8, 9]. The study of mechanically malleable organic crystalline materials is greatly captivated by it [10, 11]. Organic optoelectronics is only one example of the numerous possible applications for flexible organic crystalline materials [12-16]. Mechanical control devices, [17-19] engineered biosensors, [17] innovative biomimetics [17, 18, 20, 21], as well as the pharmaceutical sector [22, 23]. The capacity to react to environmental factors, including light, pressure, heat, solvent, and humidity, is shown by pliable organic crystals. After this reaction, there are observable morphological and motor modifications, like swaying, leaping, self-healing, swimming, jumping, curling, and twisting [6, 7, 17, 24]. Depending on whether the initial form can be regained, mechanical buckling is usually categorized as either elastic or plastic [7, 10, 11, 25, 26]. Elastic deformation is typically caused by the corrugated and/or interlocking arrangement of particles and the existence of weak and diffusing intermolecular contacts, known as "structural buffering" [26-28]. Plastic deformation relies heavily on the presence of active slip planes and the anisotropic arrangement of crystals, which involves both strong and weak contacts that are perpendicular to each other [25-28]. The crystal packing, which is determined by non-covalent intermolecular interactions, including halogen bonds, hydrogen bonds, π - π stacking, and van der Waals forces, is intimately linked to the mechanical flexibilities of organic crystals [7, 10, 11]. A great way to fully comprehend the connection between crystal structure and mechanical property is via the study of supramolecular chemistry and crystal engineering. The crystal engineering approach allows for the supramolecular self-assembly of predefined molecules, resulting in mechanical flexibilities [25, 29, 30]. Understanding the mechanical flexibility at the molecular level can assist us in designing and creating new flexible organic crystals with specific and distinctive functions and features.

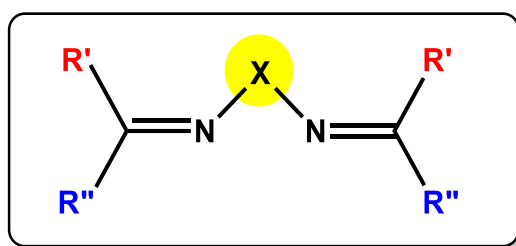
2. The Study of the Chemical Properties and Reactions of Schiff Bases

The condensation process between carbonyl compounds and primary amines produces Schiff bases. Hugo Schiff, a famous German scientist and recipient of the Nobel Prize, first identified them [31]. Structure-wise, an imine or azomethine, commonly known as a Schiff base, is a molecule that looks like an aldehyde or ketone but has an imine or azomethine group in place of the carbonyl group (C=O). Chemical compounds known as Schiff bases include nitrogen atoms connected to aryl or alkyl groups (R), but hydrogen and a carbon-nitrogen double bond serve as functional groups. Azomethane is the Schiff base. Respecting Hugo Schiff, these compounds have the following structure (Scheme 1) [32-35].



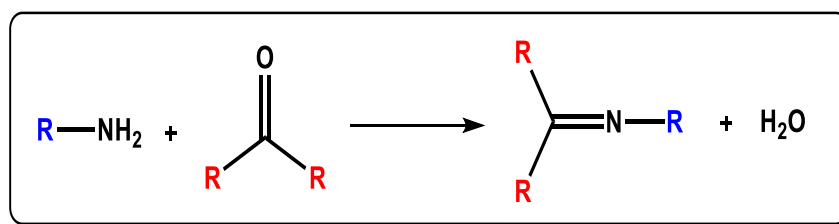
Scheme 1. The chemical structure of Schiff base [32-35].

An alkyl or phenyl group called R works to stabilize the Schiff base imine. This kind of ligand uses an aldehyde-linked group and imine nitrogen to coordinate metal ions. Chemists still make Schiff bases, and active and well-designed ligands are called "privileged ligands" [36]. Bridged Schiff's bases feature various functional groups that can change depending on the goal (Scheme 2).



Scheme 2. bridged Schiff's base [36]

R'' = phenyl or substituted phenyl, X = aryl or alkyl, and R' = H or alkyl. The stabilization of several metals in varying oxidation states by Schiff bases influences the catalytic effectiveness of these metals in various useful processes [37]. Schiff bases typically have donor groups that are NO or N₂O₂, although oxygen may also be substituted by sulfur, nitrogen, or selenium. It is often produced by condensing a primary amine with an aldehyde or ketone (Scheme 3).



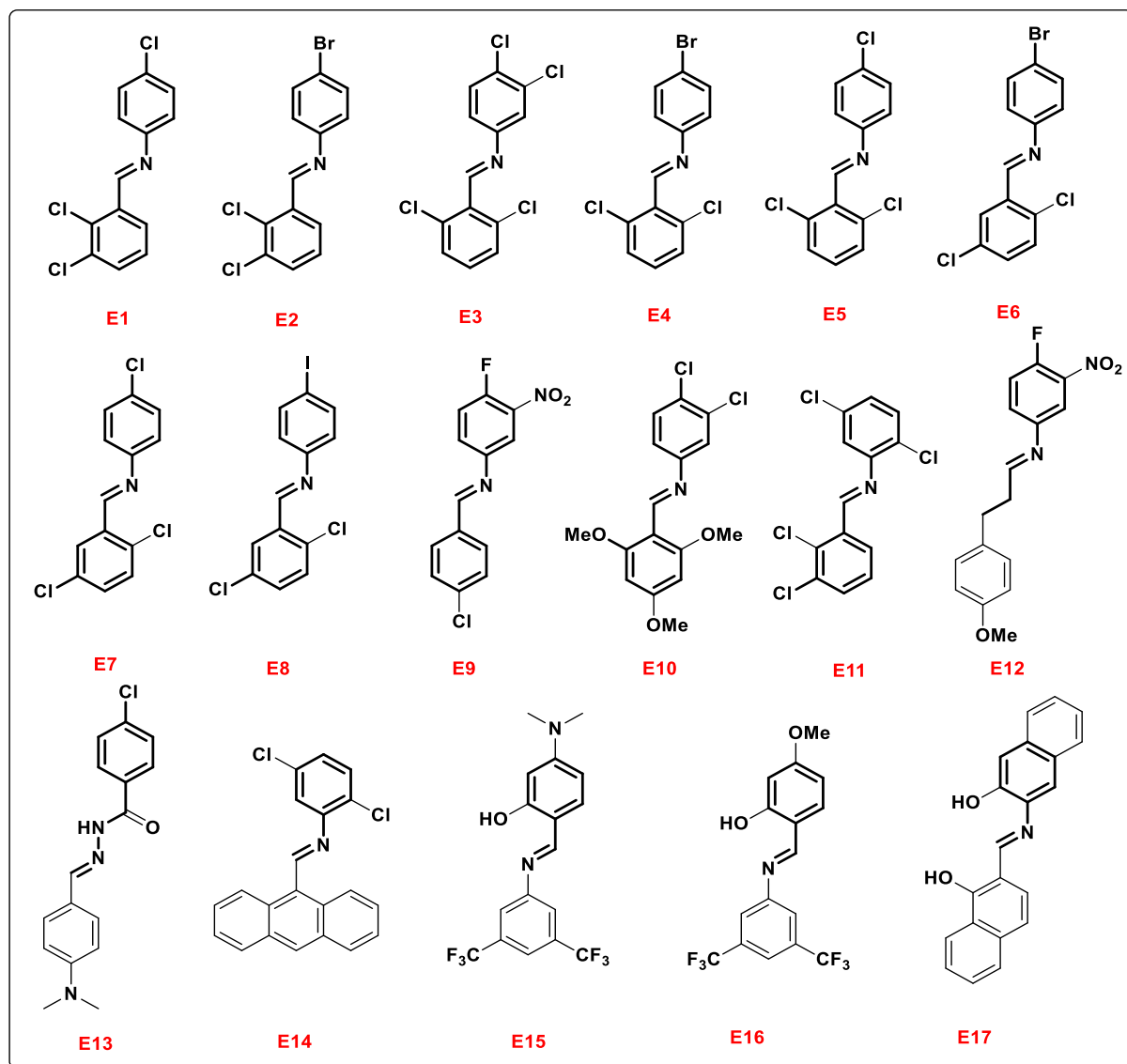
Scheme 3. Formation of Schiff base by condensation reaction processes [37].

3. Elastic Organic Crystals Formed by Schiff Bases

3.1. Schiff-base elastic organic crystals under stress.

The discovery of Elastic organic crystals on the Schiff base under mechanical tension was originally stated in 2015 by Ghosh et al. [38]. A slowly evaporating MeOH solution combined with dichlorobenzaldehyde and halogen-substituted aniline in a 1:1 stoichiometric proportion to form crystalline E1–E7 under certain climatic circumstances. According to X-ray diffraction on a single crystal (XRD) investigation, these seven Schiff base crystals were all flexible due to a common packing property: In the first place, there are three orthogonal directions of the isotropic and uniform distribution of weak/dispersive interactions (like hydrogen and halogen bonds), which have the potential to act as "structural buffering" during elastic deformation. Secondly, there are corrugated packing patterns that hinder molecular

plane sliding and long-range molecular movement. Using the Euler–Bernoulli beam-bending theory and the maximum curvature of bent crystals (Scheme 4), it was shown that all Schiff base crystals are easily bent. The maximal elastic strain was around 2%, which is higher than the majority of crystalline materials (0.5%). Three-point bending experiments determined this [38].



Scheme 4. The stress-induced Schiff base elastic organic crystals are presented in sequential sequence according to their chemical structures [38].

The three elastic crystals (E8–E10) with Schiff bases were produced using the previously mentioned synthetic method and have similar structural characteristics (Isotropic weak/dispersive connections and corrugated packing with interlocking structures) [39]. Their elastic responses were measured using three-point bending and nanoindentation. Since E9 had a higher number of variable weak interactions (C–H···O, C–H···O), it was more flexible than E8, whose typical values were 12.76 ± 0.82 GPa and 226 ± 12 MPa for E8, 8.43 ± 0.31 GPa and 291 ± 30 MPa for E9, and 12.54 ± 0.33 GPa and 367 ± 25 MPa for E10). It also had a reduced elastic modulus (E) and moderate hardness (H). Using a similar technique, several novel halogenated Schiff base crystals were created to investigate molecular architectures, intermolecular interactions, and mechanical characteristics. The only elastically bendable materials were the needle-shaped E11 and E12, which crystallized in the orthorhombic P212121 and monoclinic Pc space groups [40]. Probably, a combination of weak/dispersive

connections and isotropic crisscross packing caused the elastic flexibility. Since the former had distinct weak interactions (stronger C···N, O, and F in E12; more malleable Cl···Cl and C—H···Cl in E11), Compared to the latter, it was harder and stiffer, with elastic recovery (about 80% for E11 and 70% for E12) (Figure 1). The crystal mechanical characteristics of molecules are affected by packing features, small changes in molecule structures, and non-covalent intermolecular interactions. Similar to E13 based on hydrazides, solvable molecules also control crystal mechanical and photophysical properties [41].

The polymorph E13b, deposited at 200°C, exhibited strong green emission at 518 nm ($\Phi F = 0.19$) due to its planar conformation with smaller dihedral angles (23.68° and 63.99°) compared to E13a's greater dihedral shape in a twisted manner angles (23.68° and 63.99°). E13a, grown in dichloromethane/hexane solution, showed weak luminescence at 449 nm (quantum yield $\Phi F = 0.03$). Because of the differing packing types, the latter could not be bent, while the former possessed considerable flexibility. In contrast to E13a at E13b, the molecular chains were generated by adjacent molecules forming hydrogen bonds C=O···H—N along the c axis. This led to heterogeneous stacking and prevented elastic bending.

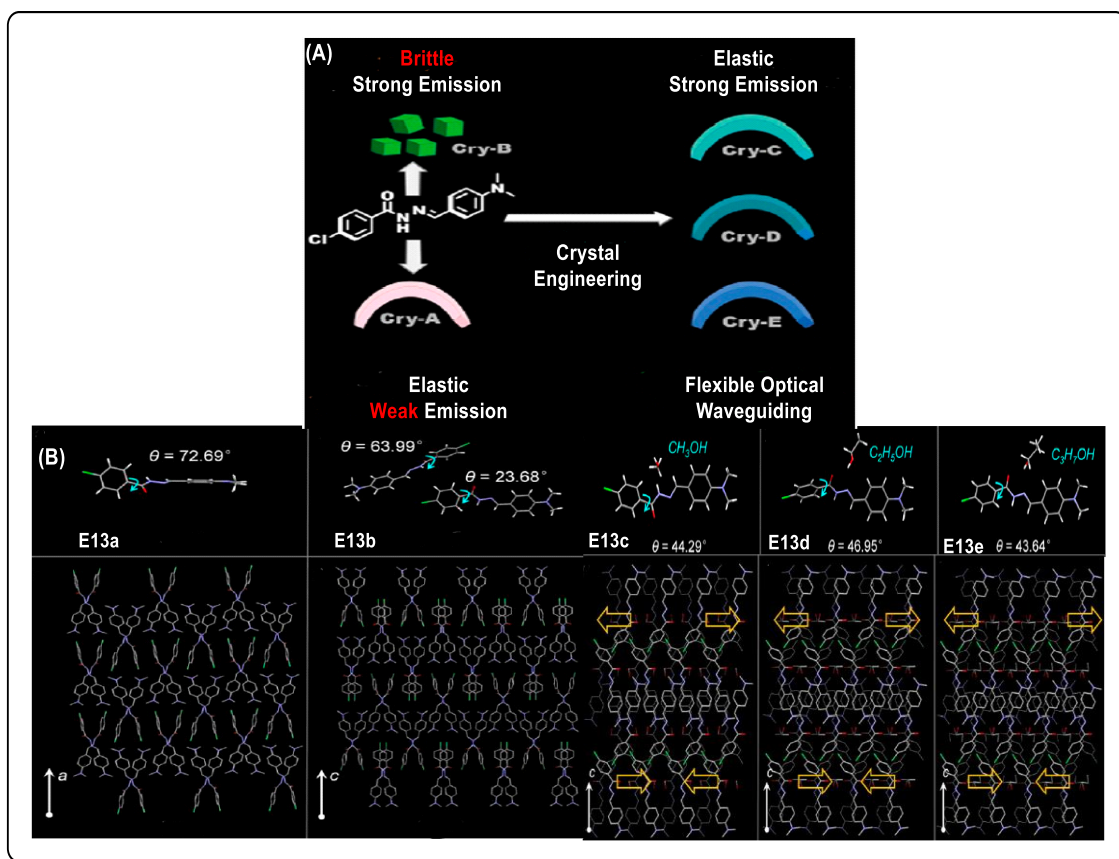


Figure 1. (A) Schematic of E13a-E13e crystals' optical and mechanical characteristics; (B) E13a-E13e molecular conformations and crystal packing. The American Chemical Society owns the copyright for the year 2020 [41].

Three solvated crystals (E13c-E13e) were created via solution diffusion using dichloromethane and several alcohols (methanol, ethanol, and propanol) by combining the benefits of two solvent-free polymorphs. These crystals exhibited good elasticity and bright emission [41]. Intense hydrogen bonding allowed solvent molecules to connect with hydrazide-based E13, creating limitless molecular chains with similar packing patterns and buffering and spreading energy during bending. E13c, E13d, and E13e crystals displayed strong emissions at 473, 465, and 461 nm, respectively, with significantly higher quantum yields ($\Phi F = 0.48, 0.55,$

and 0.59, respectively) compared to E13a. Including solvent molecules creates intensive hydrogen bonds that make crystal conformations planar, increasing fluorescence intensities and improving release behaviors. The solvated crystals show potential as pliable optical waveguides, with straight crystals with 0.149, 0.147, and 0.089 dB mm⁻¹ optical loss coefficients and bending crystals with optical loss values of 0.167, 0.166, and 0.093. Thus, crystal mechanical and photophysical characteristics depend on molecular conformations and packing modes. Because polymorphs exhibit diverse non-covalent interactions, molecular conformations, and crystal packings, they are useful tools for tuning molecular crystal characteristics such as mechanical, optical, granularity, permeability, tablet ability, and solubility. Developed from hexane, anthracene E14a/E14b polymorphs Schiff bases [42]. The crystal packings, the interpenetrating ladder-type network structure, and the isotropic packing with the crisscross arrangement of neighboring tapes were consistent with the block-shaped, elastically bending E14a and the brittle, block-shaped E14b. By modifying the crystallization conditions, the two salicylaldehyde aniline derivatives, E15a and E16a, as well as their polymorphs, E15b and E16b, were also generated [43]; its mechanical and photophysical properties are diverse. E15a released orange at 590 nm and yellow by E15b at 579 nm. More $\pi\cdots\pi$ interactions in E15a than in E15b resulted in the redshift. At 538 nm, E16a's emission spectra displayed strong yellow-green fluorescence. The acquisition of E16b was hindered by its changed molecular structure, which caused photochromic events. Mechanical behavior revealed that because of isotropic packing and greater $\pi\cdots\pi$ interactions, E15a was more elastic than E15b. Because of their separated layers, E16a and E16b exhibited flexibility and brittleness, respectively, whereas E16b's extremely deformed conformation limited molecular mobility and slippages. Notably, several tiny organic compounds with Schiff bases exhibit significant properties in both the solution and crystalline solid states, such as ion sensing [44]. Using a fluorescent probe that allows one to see live cells [45]. Consider the example of E17, which is derived from naphthalene [45]; Powder XRD and Raman investigations revealed elastic bending of the crystal due to intermolecular C—H $\cdots\pi$ interactions and O—H \cdots O hydrogen bonds. E17's two-photon excitation makes it a photo-stable, non-cytotoxic fluorescent probe that may be used to mark tumor spheroids and living cells mitochondria in solution (Figure 2). This discovery has major implications for mitochondrial disease diagnosis and therapy.

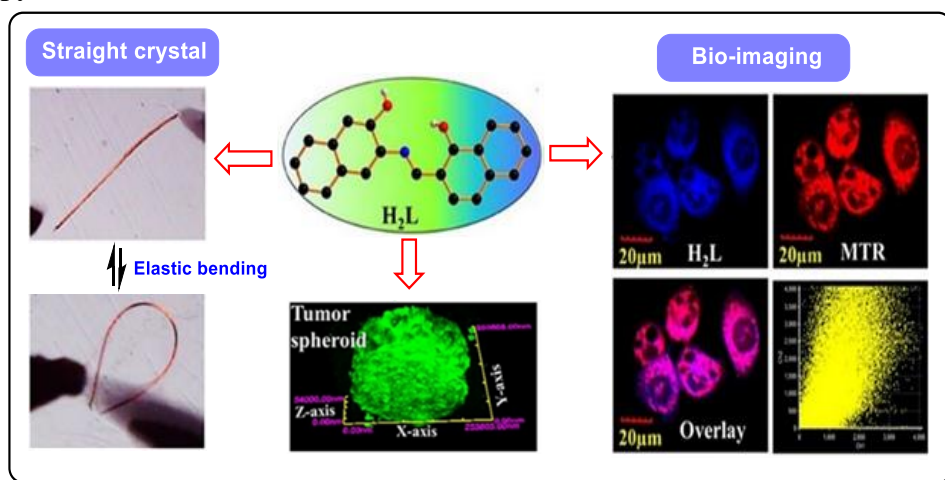


Figure 2. Schiff base E17 labels living cells and tumor spheroids' mitochondria with elastic flexibility and fluorescent probe. Reproduced with permission: The American Chemical Society, 2019 [45].

3.2. Elastic organic crystals with Schiff base functionality for optoelectronic devices.

Flexible optoelectronic devices may use elastic organic crystals based on Schiff bases as well as optical waveguides. Elastic E29 crystal was used as an optically transducing medium to build an elastic organic crystal on Schiff bases that have potential use in optical waveguides and flexible optoelectronic systems. A sandwich-type hybrid structure was constructed employing elastic E29 crystal as an optically transducing medium, Au, Ag, or Cu for electrical conductivity, and poly(urethane urea) for mechanical reinforcement (Figure 3A) [46]. The hybrid materials changed shape in response to temperature and were receptive to light and electricity. (Figures 3 C, D). With silver-coated crystals, electrical conductivity reached 21.0 S. The hybrid materials exhibited temperature-dependent deformation and were amenable to light and electricity (Figures 3C, D). Electrical conductivity with silver-coated crystals achieved $21.0 \text{ S } \mu\text{m}^{-1}$ and decreased by less than 1% during 10,000 heat cycles. The trilayer hybrid elements coated in gold exhibit rapid temperature response, exhibiting curling or uncurling in 0.2 seconds, with recovery rates of $1458.3^\circ \text{ s}^{-1}$ and deformation rates of $2187.5^\circ \text{ s}^{-1}$, correspondingly (Figure 3B). These conductive flexible cross-organic crystals have the mechanical and electrical resilience to be used in flexible organic optoelectronic applications such as thermal switching of optical/electrical circuits.

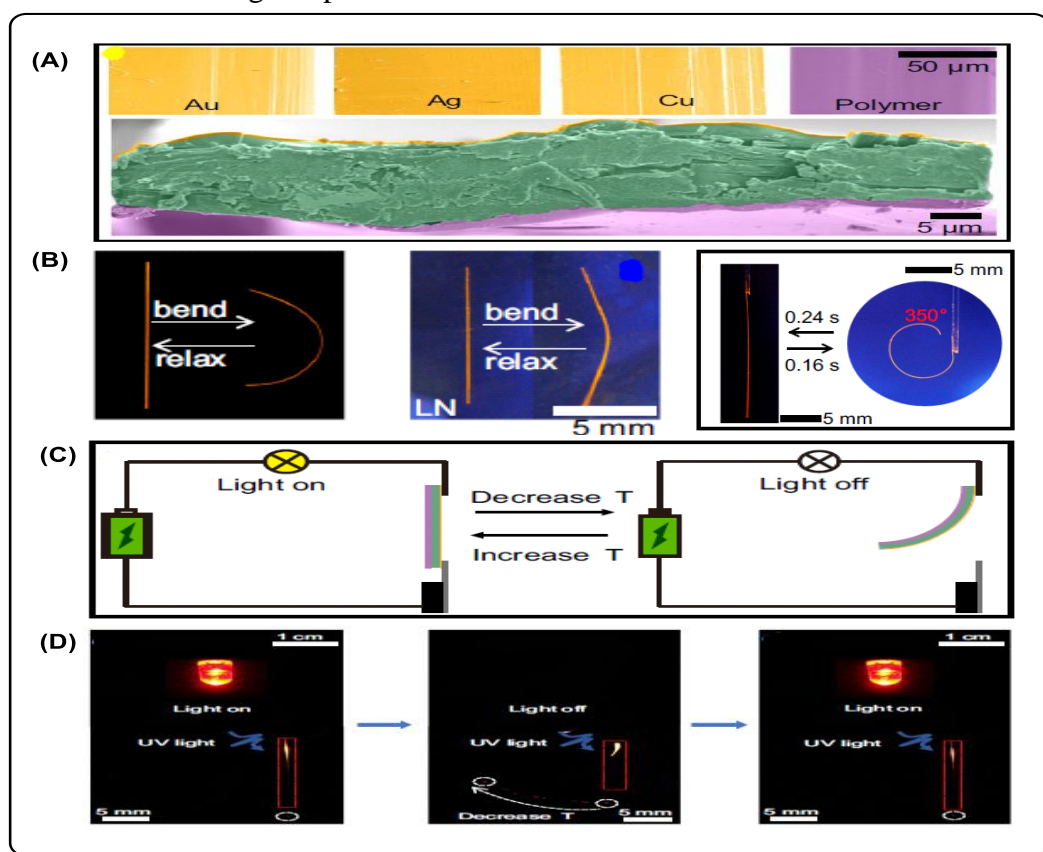


Figure 3. (A) Scanning electron microscopy (SEM) pictures with false colors that show the cross sections and surface of a hybrid sandwich structure. Metals (Au, Ag, or Cu) that function as electrically conducting components make up this structure: a polymer known as poly(urethane urea) and an elastic crystal known as E29. The crystal E29 is seen in the pictures with green highlights; (B) Pictures of bent E29 crystals produced by compression temperature-neutral (left) and in liquid nitrogen at the center, together with the instantaneous reaction of the trilayer hybrid element (Au/E29/polymer) coated in gold throughout the heating and cooling processes (right). (C) Schematic diagram showing how to use a hybrid switching element to perform heat switching in an electrical circuit. (D) A photo of the physical setup shows a mix of polymer, gold, and E29 materials, as well as a circuit that transfers signals both optically and electrically [46].

For molecular logic gates in organic electronic devices, stimulus-responsive materials represent a specialized type that may generate multiple readouts in response to external stimuli. Naphthalidenimine-based Schiff base E33's mechanical characteristics were modulated by many stimuli by Marandi et al. [47]. With a strain load, the E33 crystal demonstrated considerable elasticity, around 0.6%, due to its practically planar geometry and corrugated crystallographic packing formed *via* intramolecular hydrogen bonding between the azomethine nitrogen and hydroxyl group. Simple chemical treatments, such as attaching metal ions (like Cu(II)) The addition of acid vapors, such as glacial acetic acid, to the crystals has the potential to alter their mechanical characteristics, causing them to go from having moderate elasticity to having brittleness or high elasticity. High-resolution XPS and scanning electron microscopy, in conjunction with energy-dispersive X-ray analysis, were used to validate the binding of Cu(II) ions and the protonation of imine nitrogen atoms. Crystal multi-stimuli responses were utilized as outputs in an encoded molecular logic circuit, while chemical treatments were used as inputs in the mechanism.

3.3. Schiff base elastic organic crystals for biomimetic soft robots.

The mechanical softness inherent in most biological tissues has prompted research into artificial soft materials for biomimetic soft robots. To control mechanical properties and mimic biological processes, certain polymers with specific functions may deposit onto elastic organic crystal surfaces. Centimeter-long E29 crystals were mechanically coupled to elastic organic crystals and hyperresponsive and thermally responsive polymers to generate an interlayer adhesion-improving combination of PSS and PDDA on all sides. Subsequently, one of the broadsides was covered with a combination of a common hygroscopic PVA, PSS for reinforcement of humidity sensitivity, and glutaraldehyde for cross-linking [48]. The coated crystals were very durable and cyclable because water molecules were repeatedly adsorbed onto polymers and then released via hydrogen-bonding interactions (Figure 4). The crystals were also sensitive to temperature and humidity.

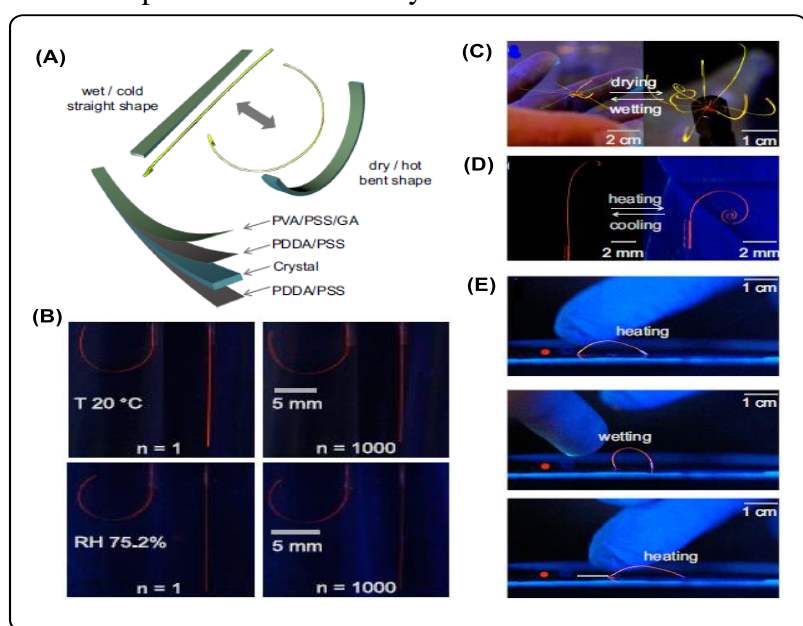
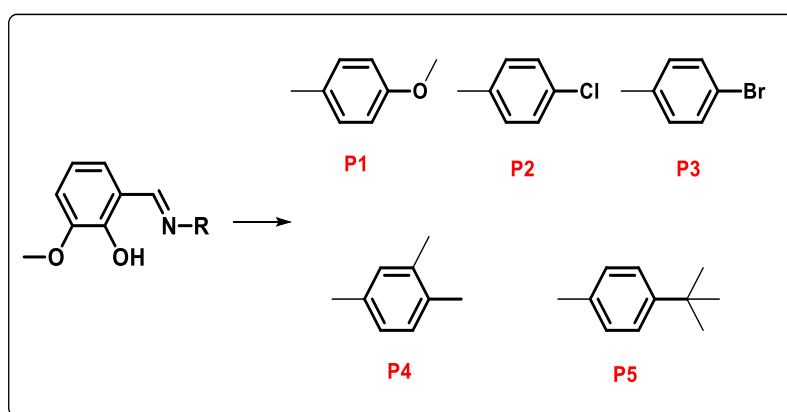


Figure 4. (A) Figure A shows the layered structure of hybrid materials consisting of polymers and elastic organic crystal E29. After humidity and temperature cycles 1 and 1000, hybrid materials with conventional and bent forms are photographed. (C) Polymer–crystal hybrid soft robots: Artificial rain lily “inflorescence” ; (D) A temperature-curling plant tendril model. “Walking” is a hybrid structure [48].

The resultant hybrid dynamic components may undergo reversible deformations that convert momentum into large-scale movements. This concept served as inspiration for the development of real-life soft robots, as seen in Figures 4C-E [48]. The differential strain at phase boundaries caused organic crystal–polymer hybrid components to imitate plant tendrils and inflorescences. After being subjected to humidity and heat, rudimentary walking robots could walk on a smooth silicon wafer. Polymers' hygro-responsive and thermally responsive capabilities have been integrated into elastic crystals to create biomimetic molecular materials in crystalline form. Any elastic crystal that can deposit and connect to elastic crystal surfaces without dissolving may be used as the driving force to create hybrid dynamic smart components when paired with responsive polymers. Molecular crystals have found a great deal of new applications thanks to these hybrid techniques, which have also moved them closer to bionics, soft robotics, and flexible electronics.

3.3.1. Schiff base plastic organic crystals.

The number of reports of bendable organic crystals has increased, albeit the majority of these were acquired by coincidence. Studies on crystals that can be bent plastically indicate that bending happens when the portion that is bent separates into pliable strata, leading to slight changes in the arrangement of the crystal lattice. The observed phenomenon may be attributed to the reconfiguration of weak intermolecular contacts, namely π -stacking and vdW forces, in the opposing orientation [8]. Reddy et al. used crystal engineering techniques to fabricate plastically flexible crystals (P1-P3) by the integration of active slip planes into unsymmetrical vanillin-based Schiff bases containing diverse vdW groups (such as π OMe, π Cl, π Br) and exhibiting significant conformational flexibility [49]. Combining non-interfering spherical vdW groups with π -stacking aromatic groups in an orthogonal orientation resulted in the development of active slip planes with little attachment energy. This combination improved the plastic flexibility of Schiff base crystals because it preserved short-range molecular movements while bending. Schiff base compounds (P4 and P5) containing alkyl substituents (π Me or π t-Bu) did not exhibit slip planes or plastic deformation due to electronegative group interference ($\text{sp}^3\text{C}:\text{H}\cdots\text{O}$ hydrogen bonds between π OMe groups). This is likely related to the fact that these compounds did not exhibit plastic deformation. Despite weak dispersive interactions being regarded as unreliable for preset structural topologies (Scheme 5), this work demonstrated that the supramolecular design of form complementarity/synthons can manage the mechanical behaviors of ordered molecular materials.



Scheme 5. The Schiff base plastic organic crystals' chemical structures are given in the order they were discovered [49].

4. Flexible Organic Crystals Composed of Schiff Bases

4.1. Flexible organic crystals that include both elasticity and plasticity via the use of Schiff base compounds.

Applications for organic crystals that may simultaneously undergo elastic and plastic deformations would be more extensive. Although integrating elasticity and plasticity in a crystal is theoretically possible, it is difficult to execute in practice. By altering the modalities of external forces, such as twisting or bending forces, Liu et al. [50]. sought to regulate various forms of deformation by influencing the arrangement of molecules in various crystallographic orientations [50]. In order to do this, a Schiff base F1 was created using a one-step condensation procedure. This base was then crystallized by progressively evaporating a solution that included anhydrous dichloromethane and hexane [50]. Two sets of parallel bending faces in the needle-shaped crystal resulted in simultaneous elastic bending and plastic twisting. It is possible to bend twisted helical crystal elastically. Hydrogen bond breaking and rearrangement were generated by irreversible molecular slippages in the thickness and breadth directions, as well as reversible alterations in $\pi \cdots \pi$ stacking along the crystal axis. $\pi \cdots \pi$ stacking controls elastic bending during twisting and is less sensitive to small changes in π -overlap degree and interaction distances, suggesting that elasticity and plasticity go hand in hand. The plastically twistable crystal F1's polarization rotators allowed for the alteration of the crystal's light output by varying the degree of twisting (Figure 5). A novel linear and flat Schiff base molecule F2 with a π conjugation was created after methyl substitution on E27 [51], whose crystal showed reversible plastic-twisting deformations and rotations of light polarization similar to F1 while maintaining the low-temperature elasticity and optical transmission capacity of E27. Views on crystal flexibility were expanded when low-temperature-resistant 2D elastic bending and reconfigurable plastic-twisting deformations were combined into one organic crystal (Scheme 6).

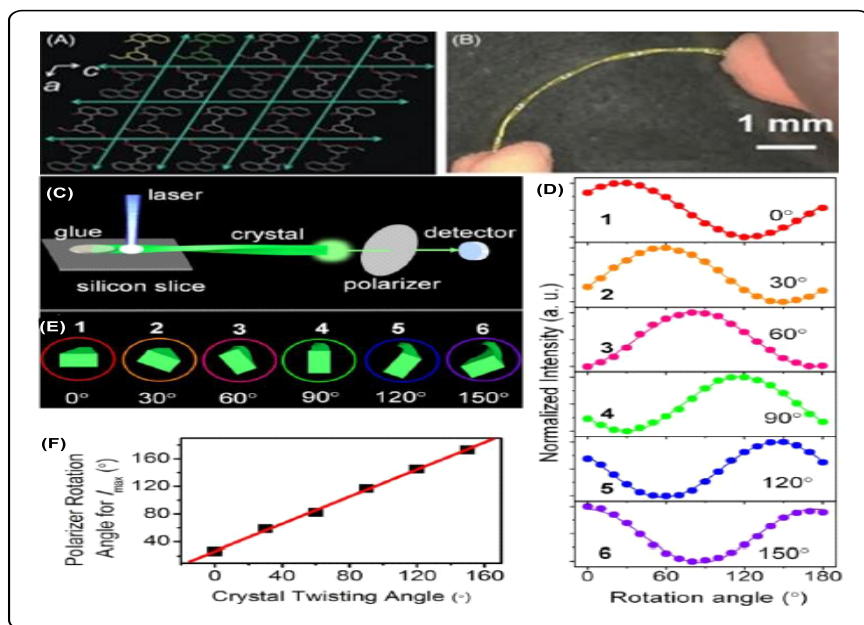
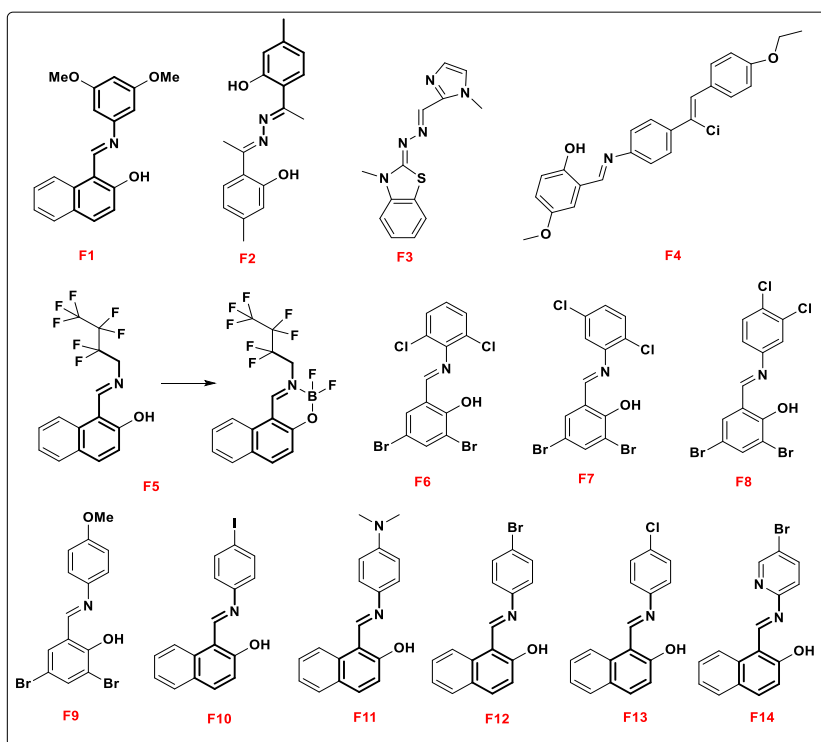


Figure 5. (A) Along the b-axis, flexible F1 molecules pack in organic crystals; (B) Twisted F1 crystal bent elastically; (C) Setup for the polarization experiment. Light intensity is measured by a charge-coupled device. For F1 crystal twisting angles, the polarizer rotation angles are used. F1 crystals twisted at different angles; (F) Maximum light intensity: a linear relationship between the polarizer rotation angle and the crystal twisty angle.

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Scheme 6. The chemical structures of the Schiff base flexible organic crystals are shown in the order in which they occur. Various pressures and external stimuli caused different mechanical deformations in F1. Based on an asymmetric azine F3, including fluorescent benzothiazole and alkaline imidazole groups [52].

Plastic bending with fluorescence intensification by UV light irradiation or acid vapor fuming, elastic bending under external stress, and reversible fluorescence switching by alternant. It was possible to produce grinding and solvent gas fuming (CH_2Cl_2 or CH_3OH). Photomechanical plastic bending was generated by molecular layer slippage and extended π -conjugation. The non-emissive crystal F3 was progressively transformed into an orange-emitting crystal with a quantum yield of 0.4% and a lifetime of 1.59 ns by UV light irradiation (Figure 6A). During processing with acid, the crystal experienced a reversible color change from green to blue (Figures 6C, D), and it bent plastically due to interactions between molecules like hydrogen bonds and protonation-induced push-pull electronic effects.

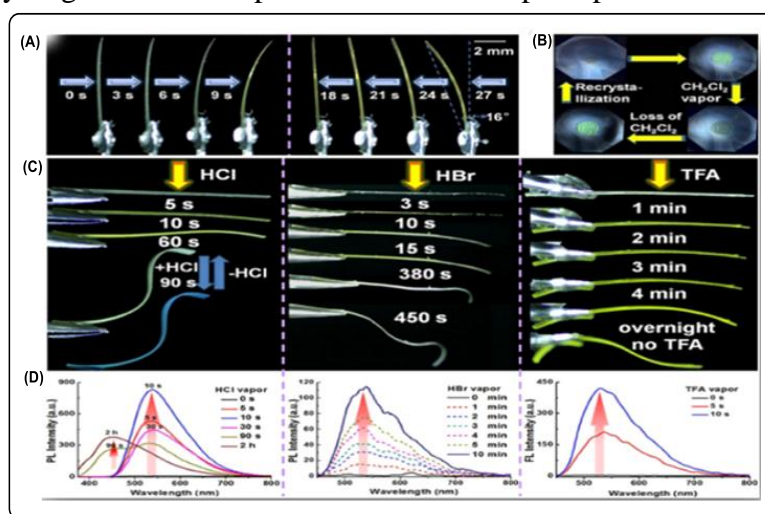


Figure 6. (A) A thin crystal F3 in the 010 plane bent photomechanically by light at 365 nm; (B) Therapy alters Crystal F3. Crystal F3 bent chemically; (C) and changed photoluminescence (PL) in HCl, HBr, and TFA acid vapor (D). Copyright 2022 by John Wiley and Sons [52].

Crystal packings, non-covalent interactions, and molecular conformations are examples of structural features that affect the mechanical characteristics of polymorphs. Three red-emitting polymorphic forms were generated by stacking π -conjugated Schiff base F4: brittle and bulky F4a stretchy tetrahydrofuran solvate F4c, which desolvated to plastic F4d with a reddish-yellow tint, and needle-like plastic F4b [53]. The structural characteristics that contribute to the distinct mechanical properties of several three-point bending and tensile tests were used to define the parameters of single-crystal X-ray diffraction (XRD) analysis. Molecules slide more easily along the crystallographic [001] and [010] axes thanks to the layered structure shown in F4b. Conversely, F4c has a crisscross configuration with vertically staggered and parallel molecular chains facilitated by intramolecular hydrogen bonding. F4b and F4c exhibited compatibility with flexible optical waveguides characterized by a minimal optical loss of 0.262 dB mm⁻¹. Changes significantly influence the mechanical and physicochemical features of crystals in their chemical composition. As hexane gradually diffused into a dichloromethane solution, sheet-like Schiff base F5 crystals and needle-like boron complex crystals with heptafluorobutyl chains and naphthalidenimine centers were seen [54]. The original F5, which lacked slip-planes and had tight packing, was brittle and non-fluorescent. The inclusion of boron produced mechanical and optical properties and formed a third aromatic ring, which promoted $\pi \cdots \pi$ stacking and C–H \cdots F interactions. The naphthalidenimine–boron complex molecules were isotropic, distributing stress uniformly during bending, with the crystallographic axes a and c serving as slip planes. Notably, due to the number of slip planes, thick boron complex crystals showed plastic bending, whereas thin ones showed elasticity. The boron complex may be used as a flexible optical waveguide in both active and passive modes since it permits a broad variety of light propagation patterns. This study illustrated how structural changes in minuscule molecules affect crystals' mechanical and optical characteristics.

4.2. Manipulating Schiff base flexible organic crystals mechanically for the purpose of creating organic photonic integrated circuits.

It is simple to characterize the optical waveguide properties of organic crystals in the millimeter to centimeter range. Micro-sized organic crystals are challenging to properly manipulate in three dimensions, which makes it difficult to downsize organic crystalline waveguides for optical micro- or nanocircuits in tiny photonic and optoelectronic devices. Using mechanical micromanipulation (mechanophotonics) techniques based on atomic force microscopy (AFM), one can create organic photonic integrated circuits by bending, slicing, cutting, lifting, aligning, transferring, and integrating organic crystals with photonic properties into optical waveguides, resonators, directional couplers, modulators, and other devices [55–57].

By reshaping and manipulating individual crystals at the microscale, Chandrasekar's group showed in 2020 how AFM could precisely regulate the light output of individual crystals [58]. An AFM cantilever tip was used to bend the 3 N-benzylideneanilines (F6–F8) to form acicular, flexible microcrystals that are pliable at any angle. After being divided from a bundle into individual crystals, these microcrystals were subsequently sliced into shorter crystals of any length. Next, a solid surface was traversed by these individual crystals (Figure 7A). These microcrystals were optical micro-waveguides that were active when stimulated by a laser. This condition determined the total transduced light's magnitude. It rearranged and reshaped little, fragile things while addressing the issue of "thick fingers". An orange-fluorescent twisted

microcrystal F9 demonstrated the technical development in non-destructive micromanipulation [59]. Hydrogen-bonding interactions (e.g., N–H···O, C–H···O, and C–H···Br) and $\pi\cdots\pi$ stack were used to achieve mechanical flexibility perpendicular to the twisted (010) and (001) planes. In both straight and bending geometries, twisted microcrystals exhibited effective fluorescence guiding, exhibiting a low optical loss of around $0.03016 \text{ dB } \mu\text{m}^{-1}$. The output signal's linear polarization was rotated by mechanically driving the optical waveguide cavity on the substrate to roll (Figures 7B–D)

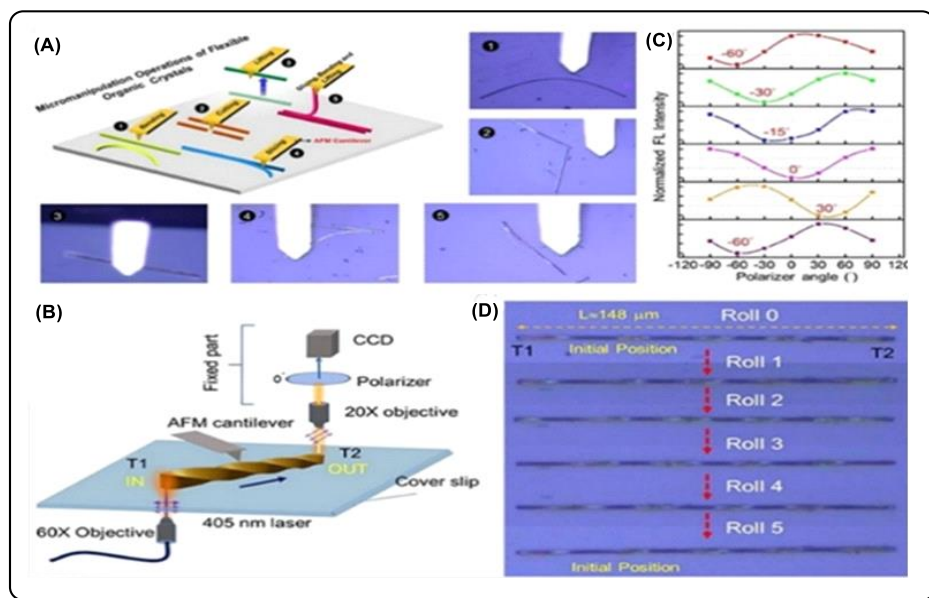


Figure 7. (A) Flexible crystals (F6–F8) undergo micromechanical processes, including bending, cutting, lifting, slicing, and bending again; (B) Experimental setup schematic showing micromechanically driven twisted-crystal waveguide polarization rotor; (C) 148 μm long twisted-crystal F9 optical photographs taken before and after rolling (R); (D) Before and after each roll, the FL intensity at terminal T2 for input from terminal T1 via polarizer angle shift (-90° to $+90^\circ$) for crystal F9. Authorization: Copyright 2020–2022, Wiley & Sons, Inc [58, 59].

The naphthalideneaniline Schiff base system (F10) used in this investigation demonstrated strong mechanical flexibility and vibrant green fluorescence, which were most likely caused by $\pi\cdots\pi$ interactions and intermolecular hydrogen bonds. F10 microcrystals could transmit fluorescence in both straight and bending geometries. Microcrystals were given pseudo-plasticity because of the larger crystal-surface adhesion energy than the crystal-shape-regaining energy. This made it possible to micromechanically create a triply bent waveguide using an AFM cantilever tip. A unique, organic photonic integrated circuit was then created by combining this waveguide with a singularly bent one on the convex region (Figure 8A). Because of the fluorescence signal's self-absorbance in the $\approx 475\text{--}550 \text{ nm}$ region, the circuit produced narrowband optical signals with tunable split ratios and direction-specific and excitation-position-dependent long-pass filters.

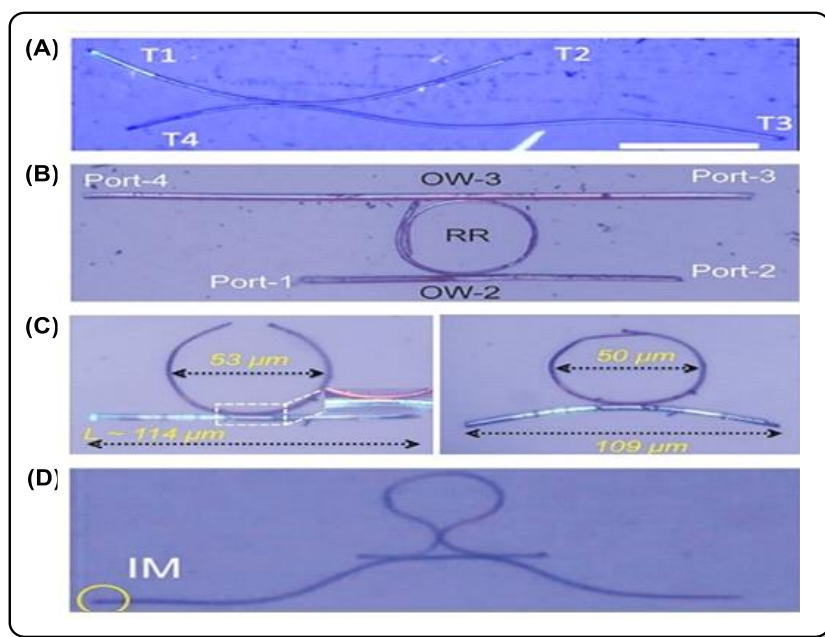


Figure 8. The flexible organic microcrystals F10, F11, F11, F12, and F14 make up the microscale organic photonic integrated circuits. Permissioned duplication: 2021 and 2022 are the copyright years. Sons of John Wiley [60-63].

Aniline structural alterations resulted in the formation of Comprising organic microcrystals that are flexible (F11–F14) [16, 61-64]; these Reconfigurable organic photonic integrated circuits by mechanical means were created using two-hybrid or the same flexible microcrystals because the substrate's pseudo-plasticity (Figures 8B–D). These microcrystals were fabricated by precise AFM-micromanipulation and used as microscale photonic components in Ring-resonators are examples of flexible organic photonic integrated circuits [61, 64], connecting elements with specific pointing orientations [61], optical fiber loop mirror interferometers [63]. The development of microscale organic photonics relies on accurate AFM-micromanipulation methods and mechanically compliant crystals, which will enable the construction of intricate organic photonic integrated circuits capable of routing a wide range of optical signals.

5. Conclusion

In summary, the rise of flexible organic crystals has revolutionized traditional perspectives on molecular materials, challenging the notion of rigidity and fragility usually associated with them. These crystals offer significant potential in various fields, such as optoelectronics, mechanical actuators, biomimetic technologies, and pharmaceuticals. The key characteristic of mechanical flexibility, crucial for practical use, depends on how molecules arrange themselves within the crystal lattice, influenced by non-covalent interactions. Using supramolecular chemistry and crystal engineering principles, scientists can customize flexible organic crystals to exhibit desired properties. Schiff base compounds, in particular, show great promise due to their easy synthesis and favorable mechanical qualities. This review presents recent advancements in flexible organic crystals based on Schiff bases, highlighting their structural arrangements, mechanical features, and wide-ranging applications. Understanding the intricate relationship between molecular structure, crystal packing, and mechanical performance aims to encourage further exploration into these materials as versatile smart materials for the future.

Author Contributions

All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest

The authors declare no conflict of interest.

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