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Low-dimensional organic Cocrystal formation mechanism, influencing factors. A Review

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Abstract

Cocrystals are composed of two or more different molecular or ionic components, which are composed of fixed stoichiometric ratio, non-solvate, and non-salt crystalline solids. According to the definition of low-dimensional organic single crystal, low-dimensional organic Cocrystal material refers to one-dimensional and two-dimensional organic Cocrystal which is different from bulk and has a lower dimension. Co-crystals are characterized by the fact that they contain at least two components connected through non-covalent interaction. Co-crystals range in electronic properties from insulators to superconductors. Low-dimensional organic Cocrystal materials show very interesting physical and chemical phenomena and many application prospects. Still, scientists face unquiet problems with organic Cocrystals structures, that's why In this review, we briefly explain the size range of organic Cocrystal, controlling structures of organic cocrystals, mainly introducing the relevant knowledge of low-dimensional Cocrystal, and following factors affecting the construction of Cocrystal, the Cocrystal mechanism or preparation strategy and Stimulating deformation so on, through theory and library research, this theme is intended to solve problems. We also focus on the Cocrystal formation mechanism, non-covalent bond interaction, and external stimuli (such as heating, light, or external force) which show a remarkable change in Cocrystal morphology caused by the change in the molecular composition of the crystal after compression. The study illustrates that organic Cocrystals can link changes in the geometric structure of molecules in the molecular world to the macroscopic motion of the material and perform mechanical work in the macroscopic system.

Keywords: low dimensional organic co-crystal, preparation, change the morphology, stimulating deformation, affecting factors, and applications.

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1. Introduction

Organic co-crystal is one of the most active fields of modern solid-state chemistry. It is a new type of organic functional material that has shown its potential applicability in various fields, including electronics, electroluminescent devices, photonics, and pharmaceutics Cocrystal refers to a class of crystals formed by the combination of an active component (host) and a Cocrystal formation (guest) through intermolecular interaction, or a variety of non-covalent bonds and interactions related optoelectronic devices has also been developed by leaps and bounds, involving many aspects of the organic optoelectronic field[1],[2],[3]. The low dimensional co- crystals are those types of co-crystals, which consist of one-dimensional and two-dimensional. These kinds of low-dimensional co-crystals are made up of atomic and ionic molecules which consist of non-covalent bonds, van der Wall force-s, and π-π staking[4],[5]. The other substances used in specific applications require comprehensive mapping of the "crystal space" associated with polymorphism[6]. Sometimes this definition is extended to specify that these components are pure solids in the presence of environmental conditions. It has been shown that different crystal forms exhibit a variety of physical and chemical properties, affecting how solid materials are applied and utilized. This study influences both basic science and practical goals[7]. Furthermore, the authors discuss different crystal forms of the same molecule, such as monocrystals, solvate-hydrate, molecular salts, and co-crystals (when the same molecule can crystallize together with a variety of co-crystal formers) and multi-functional solid, liquid crystal material[8],[9].

In theory, the organic Cocrystal molecular units can be assembled at any molar ratio, but, in the existing Cocrystal system, the most common are two constituent units, and the molecular molar ratio is 1:1[10]. There are three main mechanisms which having common in the formation of intermediate phases with higher mobility and energy than the initial crystalline species these are the liquid phase, gas phase, and solid phase[11].

In co-crystalline systems, supramolecular Synthesis is the basic concept of crystal engineering and plays an important role. An organic co-crystal has been proposed since 178 years ago as a reliable mode of intermolecular interaction that can be used to make supramolecular functional materials[12]. Another crystal engineering strategy uses electron donors (D) and receptors (A) as building blocks. Molecular structure determines the electrical and optical properties of semiconductors[13]. Organic donor (D) - acceptor (A) co-crystals have attracted increasing interest. Paul Pfeiffer published the OrganischeMolekulverbindungen in 1922, for a long time, Co-crystal did not attract people's attention. After that many scientists put forward their views on Cocrystal. Until 2012, experts and scholars from all over the world in this field of co-crystals gathered information about the scope of Co-crystals which was determined through discussion in the form of assembly[14],[15], and described many Co-crystals that were discovered at the end of the 19th and beginning of the 20th centuries; those were composed of organic components. Furthermore, the growth method significantly affects the performance of co-crystals, and further affects the performance of devices based on co-crystals, co-crystals have excellent photoelectric response performance, making them useful for future photodetectors[16].

There are also more Co-crystal based on hydrogen bonds[17],[18], which provide an excellent photoelectric response and allow for long-distance exciton transfer when combined with the DA receptor. As a result, this study not only provides a new type of white light made from room-temperature phosphorescent RTP co-crystals, but also offers how molecular structure interacts by stacking mode, and remarkable change in Co-crystal morphology after compression[19], the structure configuration and molecular composition of the co-crystal changed due to recent efforts of scientists, giving the new idea of organic co-crystals materials it is composed of p-type and n-type receptors which showing the bipolar charge transport properties. That is an excellent idea for improving bipolar organic semiconductor materials[20],[25]. In the emerging characteristics and applications of organic co-crystal, along with conductivity and field effect transistor (FET). As organic co-crystals became a hot topic, most research focused on their electrical properties (insulators, semiconductors, and superconductors) and magnetic properties[8]. In addition, the dielectric material is a kind of insulator that can be electrodes under the condition of the electric field. Since organic molecules in crystals are often strictly limited by dynamic processes, they cannot be used as dielectric materials. However, the Cocrystal with weak intermolecular charge transfer has certain degrees of freedom in the dynamic process, which is conducive to the formation of dielectric response[26],[27].

A Nobel Prize winner J.M Lehn proposed some factors affecting Cocrystal construction are introduced with thermodynamics and kinetics, supramolecular properties and having formation mechanism, and non-covalent bond interaction and external stimuli (such as heating, light or external force): This remarkable change in Cocrystal morphology is caused by the change in the molecular composition of the Co-crystal after compression [19].In recent years the growth of molecular co-crystals has been an amazing and useful phenomenon which are devolved by micro spacing in the air sublimation method. This method comprises the morphology control and essential structure properties. This method was applied on polycyclic aromatic hydrocarbon (PAH) or 1-2-4-5 tetracynobenzene (TCNB) which were grown directly on the substrate by changing the morphology from 1D to needle-like 2Dplate–like structure cocrystal. The scientific observations reveal that the crystallization process depends upon the competition between the kinetic proposed crystal and the force of thermodynamics. The growth technique of crystal has a separate demand for changing the morphology of organic co-crystals in many applications[28].

In this review, we focused on the assembly of the cocrystal formation and explained briefly the strategy of a new class of co-crystal named low dimensional co-crystal and its simulated factors such as thermal light and external force. We reported the factor that influences the low dimensional co-crystal such as thermodynamic and kinetic. Furthermore, we briefly discussed the mechanism and changing morphology of low-dimension co-crystals which opened a new era for future research in this field.

2. Methodology

2.1 Preparation Strategy of Low-dimensional Co-crystals

There was a lot of information related to co-crystals in the different chemical literature[29]. Organic Co-crystals combine with two or more different components in the same lattice through intermolecular interaction, showing novel multifunctional properties, such as bipolar charge transfer, nonlinear optics, and white light emission. Co-crystallized organic molecules with alkali, alkaline earth salt, inorganic acid, and halogen include haloquinone. The majority of organic co-crystals contain aromatic compounds, including dinitro or Trinitron. It is interesting to note that many co-crystals were discovered by different advanced screening techniques in the 1900 century, some of which were interestingly discovered by scientists. Designed co-crystals with appropriate physical and chemical properties require an understanding of intermolecular interaction and its influence on crystal stacking[30],[31].

The low-dimensional cocrystal process of synthesis and its mechanism resulted in the micro spacing being very important for morphology which governed the growth of Co-crystals in various contrivance situations. To investigate how the process of sublimation controls the morphology of the grown low-dimensional organic cocrystals. It was investigated that the process of preparation and its observations reveal that the hot stage fluorescence microscope (Figure, 1) shows the successive circumstances that arose on both bottom substrates during the growth process of the low dimensional organic cocrystal respectively [28].

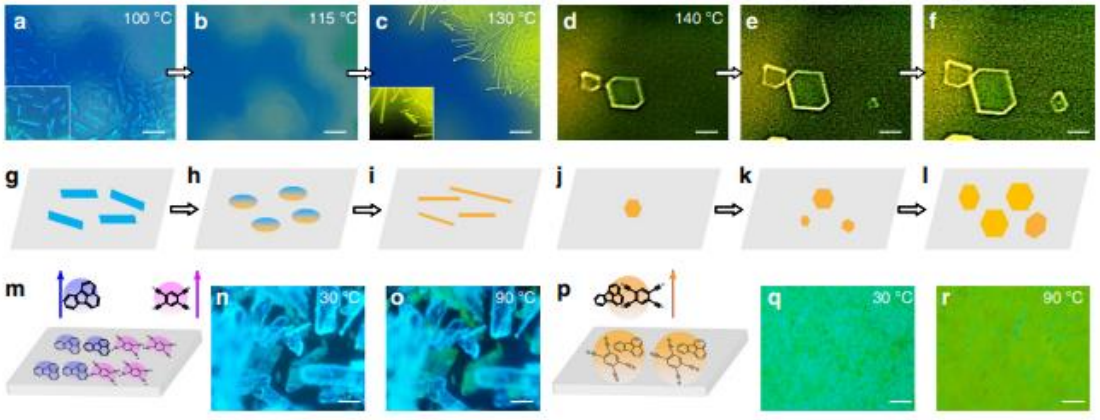


Figure 1. Real-time observation of the crystal formation process of 1D and 2D FTCs via a hot-stage fluorescence microscope. The process observed on the top quartz substrates after heating to 100 °C (a), 115 °C (b), 130 °C (c), 140 °C for 5 min (d), 10 min (e), and 15 min (f), inset is the enlarged view. The images of starting materials on the bottom substrates before (n, q) and after (o, r) heating. The corresponding schematic outline of the crystallization mechanism in the MAS method for 1D (g–I, m) and 2D (j–l, p) FTCs. (Scale bar: 20 μm for d–f, 100 μm for others). FTCs fluoranthene–TCNB cocrystals.

Different methods were used for the preparation of low-dimensional organic cocrystals. Including the solvent vaporization method that is solvent vapor annealing method and solution method, the solvent vapor annealing method is the self-assembled structure that

has been formed on the substrate exposed to a specific solvent atmosphere and subjected to a long heating annealing (strengthening) treatment [32],[33]. In the solution method, the selection of solvent, the concentration of a solution, and the hydrophilicity of substrate are the most important factors in realizing low dimensional Cocrystal. Of course, not all organic molecules are suitable for this method. According to practical experience, the method requires that different organic molecules have greater solubility and group in the same solvent. Fig 2a 2d Diffusion method [34], fig 2C The other method used is the hot saturated solution and cooling method in that method the solution was heated to the saturated state and cooled down which decreased the solubility of the solvent which resulted in precipitating the crystal and mixed with the hot two solutions and then evaporated after that the solvent was slowly reduced to room temperature, and then the solvent is poured out and washed to obtain the Cocrystal of the two organic molecules[35]. Fig 2b Professor Shuit-Tong Lee, Professor Liao Liangsheng, and their research group in the year 2012 proposed a reprecipitation method. This method explained that two organic molecules were mixed and dissolved by certain molar ratios but usually used 1:1 ratios and injected into a large number of partial soluble solvents (partial soluble solvents can be mixed with the solvent) so that good solvents and partial soluble solvents have a very good mixture and organic molecules under influence of a large number of solvent atmosphere, the solubility is rapidly reduced, and molecular aggregation nucleation occurs[36]. In addition, the transfer method Physical Vapor Transport (PVT) method refers to the use of heating sublimation of organic molecules to form a vapor, driven by external carrier gas, from the high-temperature region to the low-temperature region [12]. Fig 2E, this kind of chemical change between solids and solids under the action of mechanical force provides an important reference for the development of new Cocrystal materials, and the reaction mechanism has also been deeply studied in the literature[10],[37] A systematic study was conducted on the influence of crystallization methods on the obtained series of organic cocrystal solid forms. A cocrystal of benzoquinone (BQ) and one of the four compounds. Monosubstituted p-halogenated phenols were prepared using chloroform as raw material through crystallization, co-sublimation, vacuum, and pure grinding processes. Co crystals with donor receptor ratios of 1:1 and 2:1 can be prepared [111].

These methods of Cocrystal have an important influence on the formation mechanism of Cocrystal and the formation mechanism of Cocrystals. Grinding cocrystallization is not a single mechanism-dominated process, but a series of mechanisms including molecular diffusion and cocrystal alloy formation. Amorphous co-crystallization and so on. In theory, the organic Cocrystal molecular units can be assembled at any molar ratio, and the stacking mode of the Cocrystal. But, in the existing Cocrystal system, the most common are two constituent units. Usually, most organic co-crystals are formed at a stoichiometric ratio of 1:1 in the D-A system. 1: 1 binary complex can be packed in two ways: one is separate (segregated) stacking, where the same molecule like a donor or acceptor is packed into its column, like -DDDD- and -AAAA- way. The other is hybrid (mixed) packing, where different molecules pack in alternative ways, modeled as –DADADA–. As well as two additional donors are inserted in the mixed-stacking column and form another additional column, respectively (-A-D-D-A-D-D-A-D-D- and -D-D-

D-)[38],[39],[40]. The above mechanisms have a common form of intermediate phases with higher mobility and higher energy than the initial crystalline species, so a small amount of liquid has little effect on the final stacking structure[41].

The non-covalent interaction between molecules is the important reason why molecules can interact in a specific way to form highly ordered crystalline materials. The non-covalent interaction between molecules affects and determines the self-assembly process and kinetics process of the Co-crystal and plays a crucial role in the properties, structures, and stability of the Co-crystal. Once a crystal lattice is formed. The interactions that communicate between molecules is controlled by various intermolecular interaction that ultimately determines the physical properties and reactivity of the crystal. Much of the current fundamental research work in crystal engineering focuses on the development of reliable supramolecular reactions which can be judged according to the frequency generated by the desired intermolecular forces and the mode of connection In many cases[42],[43],[44],[45], Common non-covalent interactions include charge transfer (CT interaction). π -π[46],[47],[48], (m-x interaction) hydrogen bonds or halogen bonds[49],[50],[51],[52],[53].

The most common intermolecular interactions in Co-crystal in charge transfer are widely present in organic molecular donors and acceptors. In charge transfer, the Co-crystal system charge is transferred from the highest occupied molecular orbital (HOMO) of the donor to the lowest unoccupied molecular orbit (LUMO) of the acceptor [38],[39],[46].

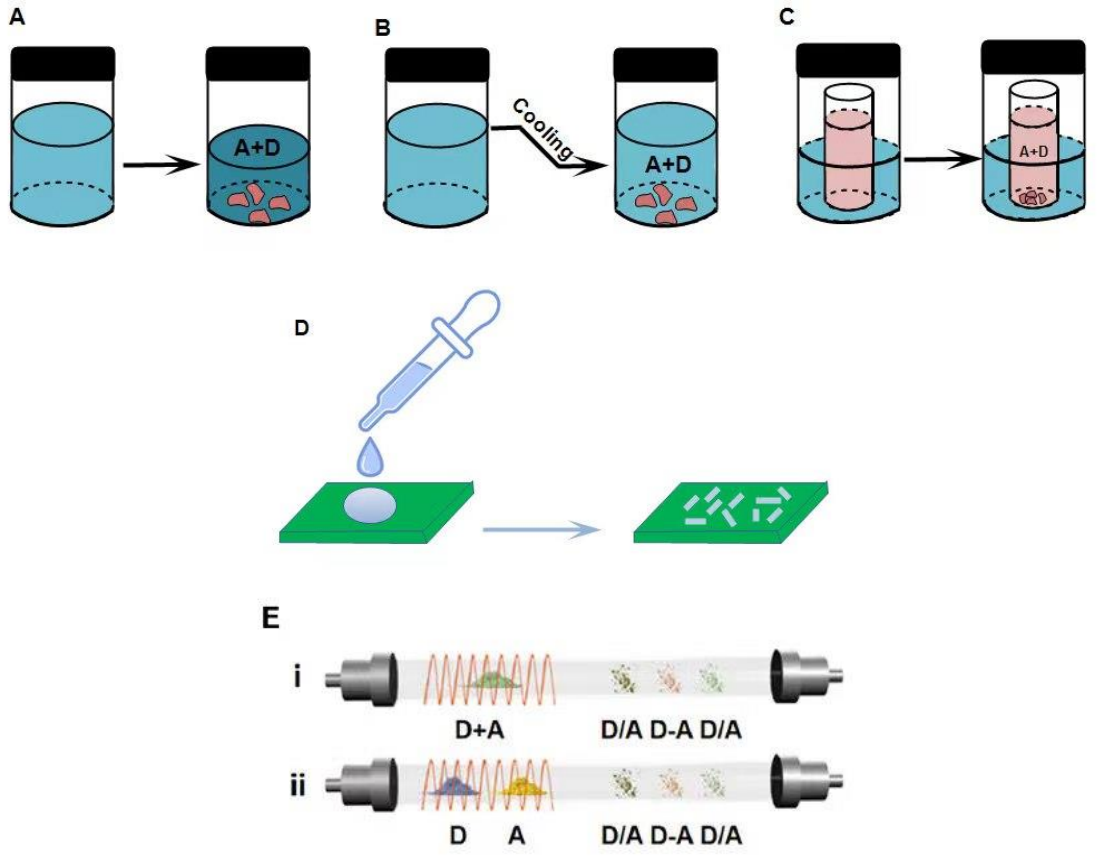


Figure 2. Schematic illustrations of the co-crystal growth processes of (A) slow evaporation, (B) slow cooling evaporation, (C) diffusion, (D) drop-casting, and (E) physical vapor transport (PVT) (Reproduced from Sun et al. (published: 09 December 2021 doi: 10.3389/fchem.2021.764628).

2.2 Size of the Organic Co-crystals

It has been demonstrated that different sizes of organic co-crystals can be obtained using these methods, including micro, Nano, small, and macro-crystals. We have explained in Table 1 that there are three main methods for effectively preparing co-crystals, including the liquid phase method, gas phase method, and solid phase method. Co-crystal properties and performance of co-crystal-based devices are greatly affected by the growth method, so it is important to select appropriate growth conditions based on the inherent characteristics of the components [6]. However specifically, the solvent evaporation method usually dissolves a certain proportion of the reaction mixture solution in a bottle and then stores it in a dark environment. With the evaporation of the solvent, the solute gradually reaches a saturated state in the solution, forms crystal nuclei driven by intermolecular interactions, and gradually precipitates out after a few days. This method usually requires donor and acceptor molecules to have similar solubility in the same solvent and is highly dependent on factors such as solvent, concentration, temperature, and other external conditions [38].

There have been reports of various types of co-crystals, based on the number of components present in the crystal structure (binary, ternary, and quaternary Co-

crystal)[54]. Consequently, more than 2000 papers have suggested that pentacene transistors with different solid phases and morphologies depend on growth conditions and/or the type of dielectric interface. A flow-enhanced crystal engineering technique for single crystal polymorphs and crystal structures[55]. In 1844, Wöhler published his first article on co-crystals. As early as 1973, John Ferraris discovered the TTF-TCNQ co-crystal (TTF, tetrathiafulvalene; TCNQ, tetracyanoquinodimethane) which had high conductivity. The growth methods have a significant impact on the co-crystal properties [6]. The table below presents a review of all the different sizes of organic co-crystals, such as micro/non-crystals, macro/big co-crystals, and small or amorphous co-crystals, which were developed using the above methods.

In summary, this systematic study was conducted on the influence of crystallization methods on obtaining different solid forms of organic co-crystals. The study involved preparing co-crystals of benzoquinone and various monosubstituted p-halogenated phenols using different processes such as crystallization, co-sublimation, vacuum, and pure grinding. The study demonstrated the preparation of co-crystals with different donor-receptor ratios. In a 1:1 binary complex, two packing modes are commonly observed. The first is separate (segregated) stacking, where identical molecules (either donor or acceptor) are packed into their respective columns. The second is hybrid (mixed) packing, where different molecules alternate in the stacking, represented. Additional donors can also be inserted into the mixed-stacking column, forming additional columns. In addition, different sizes can be achievable in organic co-crystals and introduce the liquid phase, gas phase, and solid phase methods for their preparation. It emphasizes the importance of selecting appropriate growth conditions based on the components' characteristics and provides specific details about the solvent evaporation method.

Table 1. Different Methods and Sizes of the Low-Dimension Organic Cocrystals

number	**Liquid-phase Methods:**	
1	There are three common liquid phase methods: slow evaporation, drop-casting, and diffusion method.	
	Methods	Sizes of Organic Co-crystals
1.1	liquid phase methods: slow evaporation,	The slow evaporation method is used to grow co-crystals with big sizes.
1.2	liquid phase methods: drop-casting,	The drop-casting method is used to prepare micro/ Nano co-crystals for constructing optoelectronic devices.
1.3	Liquid phase methods: diffusion method.	In this method, the solution concentration is a crucial factor affecting the micro-/nanostructures of co-crystals.
	Vapor(gas)-phase Methods:	
2	There are two types of PVT methods according to the sublimation point of the ingredients, and the other type of PVD method applies to the ingredients with significantly different sublimation points, in which the donor and the recipient are placed in two furnace areas	
	Methods	Sizes of Organic Co-crystals
2.1	PVT methods according to the sublimation points of the constituents'	two sizes micro and macro sizes
2.2	The PVT method is appropriate for constituents with significantly different sublimation points.	micro co-crystals
	Solid-phase Methods:	Sizes of Organic Co-crystals
3	Solid-phase methods produce fewer organic co-crystals than the methods mentioned above.	The products always have small sizes and irregular morphologies.

3. Result and Discussion

In this review we have discussed, the basics of organic co-crystals and briefly explained the morphology of low-dimensional co-crystals. In detail how these types of co-crystal are formed and which kinds of interactions are present in them. Furthermore, simulated factors that affect the low-dimensional co-crystal structure change their morphology, and also explain some of the basic applications of the low-dimensional co-crystal which are unique in the field of co-crystal applications.

3.1. The Factors Affecting the Construction of Cocrystal

Most organic Co-crystals are formed by intermolecular charge transfer, π-π interaction, hydrogen bonds, one or several halogen bonds, and van der Waals force. in terms of crystallographic engineering, Cocrystal is generally divided into three categories: first two-component Cocrystal composed of two organic solid molecules; second Multi-component Cocrystal formed by co-crystallization of solvents and two or more solid molecules; third Co-crystal salts of organic compounds[56],[57],[58],[59],[60],[61]. For designing of co-crystal it is necessary to understand the characteristics and molecular structure of organic acceptors and donors. The suitable and important selection of Co-crystal units plays an important role in the development of new materials. It is essential

to understand important two aspects: first to understand the stacking mode of molecules (i.e. the combining mode of cocrystal) which essentially affects the properties of solids. Secondly to understand how to aggregate, nucleate crystallize, and how different molecules recognize each other which are the influencing factors of Cocrystal construction. Furthermore, different suitable guests and hosts were designed and studied their various properties for the construction of single-component organic Co-crystals [62]. A Nobel Prize winner J.M.Lehn writes molecular chemistry which made amazing developments in covalent bond manipulation, and now the time comes that is necessary to show the power of non-covalent bonds which means the formation of Co-crystal through non-covalent bonds. Cocrystal is a highly ordered supramolecular crystalline material formed by the interaction of different molecules in a certain stoichiometric ratio through non-covalent bonds. Four factors affecting Cocrystal construction are introduced in turn: thermodynamics and kinetics, supramolecular synthesizer, Cocrystal formation mechanism, and non-covalent bond interaction [63]. The thermodynamic and kinetic factors rely on crystal engineering, its balance of molecular interactions, and space-filling effects such as chemical and geometric factors. The chemical tactic depends on the energy levels of various possible staking structures[64],[65].

In physical chemistry two important parameters are used for the thermodynamic system i.e. enthalpy and entropy of the material system these parameters are related to the measure of heat and disorder of the system respectively. When two different molecules are crystallized the enthalpy and entropy will take part in the structure change. When effective molecular and size of the component is formed the crystal packing is favorable and the enthalpy of the system is greater than entropy which increases the order of the crystal formation, when the components of the molecule mismatch then the entropy is greater than the enthalpy of the system so the binding force dominates and amorphous cocrystal is formed [11].

When there is a strong interaction between different molecules that is greater than the same molecules, it is favorable to achieve enthalpy-driven Co-crystallization[66]. Most interestingly, the phenomenon of crystallization is kinetic, so according to the kinetic phenomena the intermolecular interaction first forms a solution sealed with each and binds with isotropic force. The choice of solvent thermodynamically plays an important role in cocrystal operations and its growth. The synthesis of organic cocrystal is the basic unit that is formed by the linkage of non-covalent interactions between specific molecular influencing groups to synthesize cocrystal structure. The method of formation and mechanism of cocrystal are very important in the cocrystal structure which is reviewed in detail in the portion of the formation.

There are different methods of formation of cocrystals despite grinding cocrystallization such as molecular diffusion, cocrystal alloy and amorphous co-crystallization so on. In the above cocrystal formation methods the intermediate phases with higher mobility and energy than the initial crystalline species. The Cocrystal formed by grinding is usually thermodynamically stable, so a small amount of liquid has little effect on the final stacking structure[67],[10],[41].

3.2. Stimulating Factor and Deformation of Cocrystals

Many crystalline materials are found to exhibit different properties (i.e. ferroelectricity, electrical conductivity, and magnetism) in different crystalline phases. To understand the structural properties of Co-crystal it is important to know the polymorphs of Co-crystal and its chemistry[68],[69],[70]. However, the changes in physical and crystal size in the phase transmissions remain relatively unknown. In addition, Williams, R. M reported that Single-crystal-to-single-crystal (SCSC) phase transitions with significant dimensional changes can provide us with a good model to study the correlation between crystal internal changes and their morphology[71].

In recent years, stimulated deformation molecular crystals have received more and more attention in the scientific community, such crystals usually undergo macroscopic deformation or even mechanical movement when they are stimulated by external stimuli (such as heating, light radiation, and external force[72],[73],[74]. For performing mechanical work, scientists construct such a molecular system which having mechanical motion based on the geometry of individual molecules induced by external stimuli. For example the molecular scale movement in biological systems - Myosin is ingeniously linked to the macroscopic movement of muscles. Despite the elaborate design and synthesis of various types of molecular machines[75][76],[77]. However, the global similarity of crystal structure before and after phase transition and the local key differences (stacking mode, rotor orientation, and intermolecular force) is the origin of the self-repair properties of the single crystal Material. This study gives us a new understanding of the phase transition mechanism of molecular crystals and provides new insight for the further design and study of crystal self-repairing materials.

3.3. Light and Heat

In 2010, a Japanese scientist designed a photo-deformed organic cocrystal, in which the naphthalene substituted dithiophene molecule performs as a donor and the acceptor molecule is a perfluorinated substituted naphthalene (FN), in that organic cocrystal. After irradiation with UV (365 nm) light from the right side of the crystal, the crystal turns blue and bends away from the light source, as shown in (Figure 3a,b), the Cocrystal exhibits rapid and durable light-induced bending behavior under ultraviolet/visible light irradiation in the temperature range of 4.7 to 295 K [78]. When the lights were turned off, the bending stopped and the crystals maintained their curved shape in the dark. When the bent crystal was irradiated with visible light ($\lambda > 440$ nm) from the right, the blue color disappeared and the crystal returned to its original rectilinear shape. Figure 4a-b shows the effect of visible light illumination. The crystal is irradiated with UV light from both sides and then irradiated with visible light from the right side. At first, the crystal bends toward the light source and then becomes straight[79]. The X-ray crystallographic analysis shows that the photoinduced deformation behaviors caused by the anisotropic expansions of the crystal lattice caused by the change of the molecular shape during the photoinduced cyclization of the Cocrystal component LO molecule. The study illustrates that organic Co-crystals can link changes in the geometric structure of molecules in the molecular world to the macroscopic motion of the material and perform mechanical work in the macroscopic system [78].

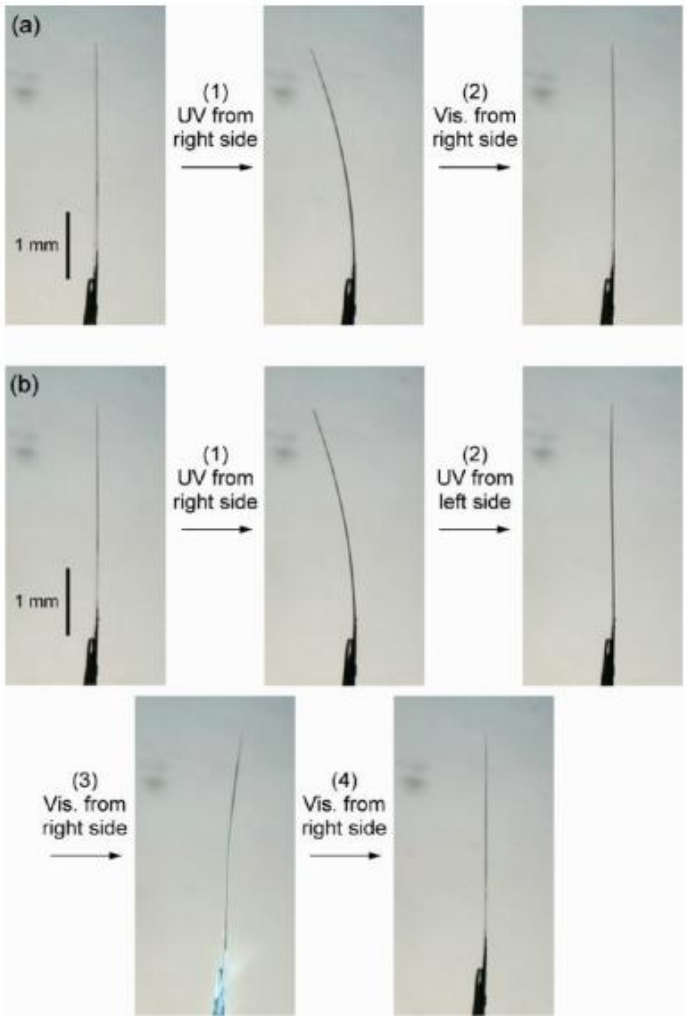


Figure 3. Reversible bending of a cocrystal of LO ·FN (3.1 mm × 0.5 mm × 20 μm) induced by irradiation with UV (Keyence UV-400 with UV-50H, 365 nm, 30 mW/cm2) and visible (Asahi Spectra MAX-302, λ > 440 nm) light. (a) (1) Upon irradiation with UV light from the right side, the crystal bent, moving away from the light source, and then (2) returned by visible irradiation. (b) The crystal (1) first bent by UV irradiation from the right side and (2) second straightened by UV irradiation from the left side. (3) Upon irradiation with visible light from the right side, at first the crystal bent two.

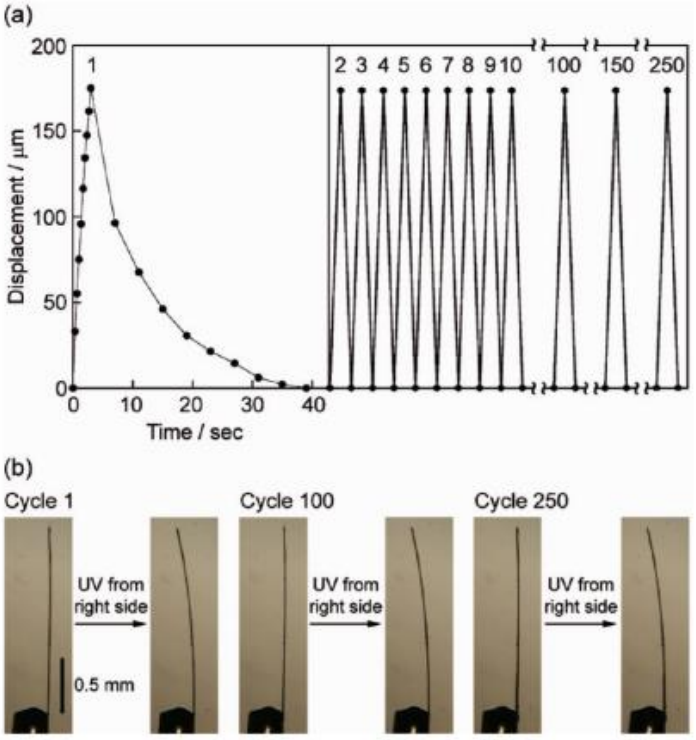


Figure 4. Reversible bending of a cocrystal of 1o ·FN (1.7 mm × 0.2 mm × 10 μm) by alternate irradiation with UV (365 nm) and visible (λ > 440 nm) light. (a) Tip displacement of the crystal during the reversible bending. (b) Photographs of the crystal in the 1st, 100th, and 250th cycles of the reversible bending.

There is limited research in the field of deformation Co-crystal but in recent times in this field, the organic cocrystal has been effectively applied to stimulate deformation in the field of molecular crystals through mechanical pressure and light heat. For example Tao Xutang of Shandong University. Constructed the Cuq2-TCNQ cocrystal composed of 8-hydroxyquinoline copper (Cuq2) and 7, 7, 8, 8-tetracyno-p-phenylenediquinone dimethylene (TCNQ) [19].

Through deformation stimuli, the remarkable change in Cocrystal morphology occurs, after compression. In (Figure 5). It was observed that the crystal II completely transformed into form I without breaking, which means that form II is metastable relative to form I at room temperature. But this transition state is for only a few seconds. From scanning microscopic studies as shown in Figure 5 it was assumed that the crystal shows the morphological change in the transition state and roughly double, and defects of the crystal are very expanded in size, the plane (001) of form II becomes (111^{-}) and form I remains same. Therefore, the authors suggest that a probable phase transition mechanism described as follows: first, a large number of nuclei generated by mechanical stimulation of the type II crystal, which leads to the molecular rearrangement that generates the phase transition; then, the construction of 2D layer is created layer by layer; finally, the complete transition induces a crystal size significant changes [80],[81].

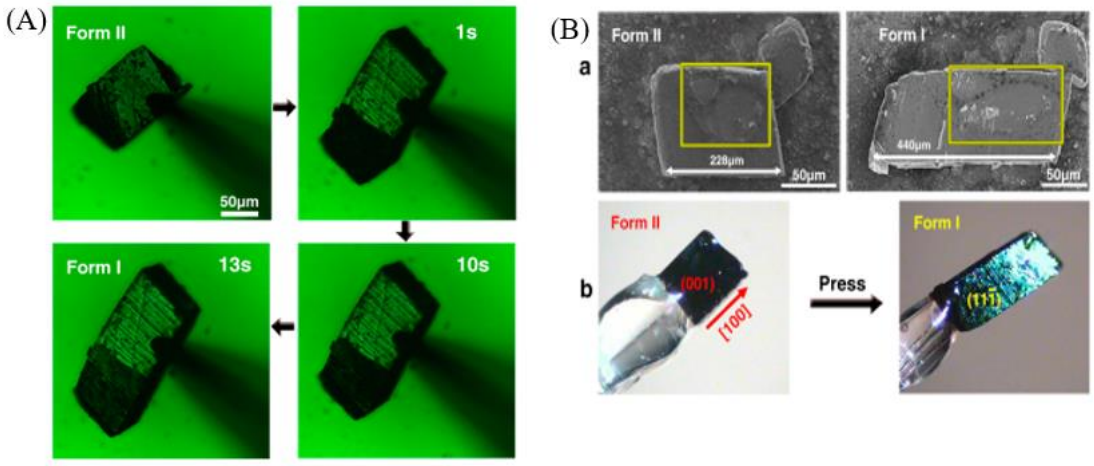


Figure 5. (A) Cuq2-TCNQ Cocrystal phase transition under pressure, (B_a) Scanning electron microscopy images of the crystal before and after phase transition, and (B_b) the changes of surface defects on the crystal.

3.5. Solvated Cocrystal

We reviewed many scientific works in which different solvents were used for the preparation of organic co-crystals and had an effective role. Hu Wenping et al. By using a slow solvent evaporation process, a red-emitting perylene-TCNB-2THF solvate co-crystal was developed. Through the gain and loss of solvent molecules, the solvated co-crystal could be reversely transformed into the gray-green non-luminescent perylene TCNB two-component co-crystal (by adsorption or removal of THF solvent). It is particularly noteworthy that the perylene TCNB two-component co-crystal gradually produces significant mechanical bending when adsorbing THF solvent molecules, as shown in Figure 6. This special stimulus-response deformation phenomenon not only exists in the stimulation process of tetrahydrofuran but also exists in the stimulation process of some organic solvents with similar molecular configurations. This change in the macroscopic state of the co-crystal caused by the change in the microscopic state inside the molecule provides an effective model for studying the deformation mechanism of the co-crystal and is expected to further expand the application range of the co-crystal in the field of intelligent response[82].

A novel co-crystal and its solvate co-crystal were designed and constructed by Jing Wang et al. Structural and electrostatic potential analysis revealed that solvation disrupted the N−H···N hydrogen bond interactions between phenothiazine (PTZ) and 1,2,4,5-tetracyanobenzene (TCNB), resulting in TCNB the molecule has a 90° rotation about the normal axis of the PTZ plane. Therefore, the D−AC−C surface close contact is enlarged, and the molecular π–π stacking interaction and intermolecular CT interaction between PTZ and TCNB are strengthened, which is further evidenced by absorption and Raman spectroscopy analysis. This study provides rare evidence for the expansion of close contacts on the C-C surface in mixed packing between D and A, which greatly facilitates the intermolecular CT interactions in the D-A co-crystal system. It also provides a deeper

understanding of the role of solvation in structure-property relationships in organic co-crystal materials [83].

Yuan Wang et al, characterized Tris(8-quinolinolato)aluminum(III) (Alq3) by polymorphic crystallization under vapor deposition and solvent evaporation, and the crystal growth of Alq3 in several organic solvents was investigated. High-quality bulk single crystals or crystalline microstructures were obtained, which depended upon different crystallization conditions. Two new crystalline solvates Alq3(HCON(CH3)2) and Alq3((CH2OH)2) were grown in N, N-dimethylformamide (DMF), and ethylene glycol (EG) were obtained. X-ray diffraction analysis revealed that their structures belong to a new polymorph with the same crystal symmetry. It demonstrates that solvated single crystals or microstructures can be used as excellent light-emitting and propagating materials in miniaturized optoelectronic devices[84].

Al-Kaysi et al, have described the reaction of single-component molecular crystal nanorods with a second species to form cocrystal nanorods. a porous anodic aluminum oxide template was used on which Single-component crystalline nanorods, composed of 9-methyl anthracene (9-MA), then the 1,2,4,5-tetracyanobenzene (TCNB) in water these templated rods were exposed which slowly diffuses into the 9-MA rods over days and 1:1 charge-transfer complex within the rods were formed, which were transformed from crystalline 9-MA into co-crystalline 9-MA/TCNB. The cocrystal nanorods are characterized by electron microscopy, X-ray diffraction, and optical spectroscopy, confirming their highly crystalline structure and the formation of the charge-transfer complex [85].

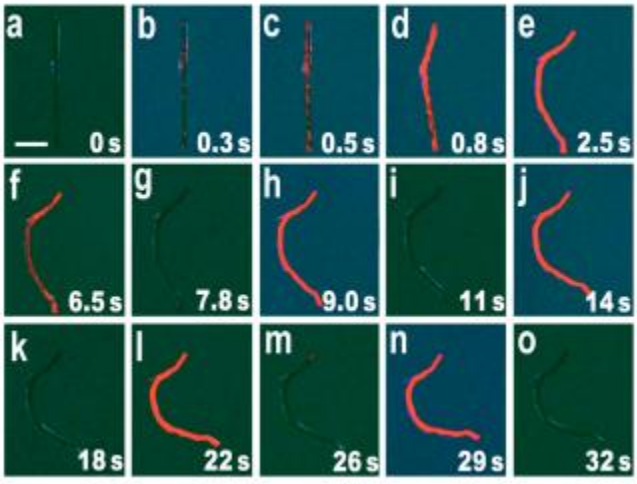


Figure 6. Reversible structural changes between two forms of Cocrystal show intent and their effect on tetrahydrofuran vapor (b)~ (e) and Color and shape changes occurring in (h-j,l,n) and as well as air [a, f, g, I,k, m,o].

The stimuli-responsive molecular crystals may also be extended to organic Co-crystal, such as remarkable changes base of TCNQ as CUQ2-TCNQ upon the mechanical stimulation. Furthermore [19], it was confirmed by IR measurement that IR bands originating from perylene donor or TCNB accepter did not alter when it was compared

with the constituent materials. However, the PXRD results reveal that the peaks and shapes are different before and after deformation. The PXRD analysis of PT2TC and PTC was also examined. When the PT2TC release the THF molecule peaks obtained were weaker from the initial as shown in Figure 6c and almost shifted as compared to those of the former one as shown in Figure 6b)[82].

3.6. Thermal Deformation Effect on Organic Co-crystal

The other authors also investigated that the thermal deformation and stimulation of organic Cocrystal shows huge changes in their structure. In addition, mechanically responsive materials exhibit remarkable macroscopic changes as a response to external stimuli, including heat, light, and mechanical force [86],[87],[88],[89]. To achieve structural deformation and stimulation, it needs careful analysis of the performance and characterization of the single crystal by getting sufficient information, such as staking patterns, molecular geometry, and key intermolecular interactions[90],[91],[92],[93]. There is various example of thermal deformation of the cocrystal such as coronene – TCNB, dipyrazolethiuram disulfide crystals, and so on [94],[95],[96]. Tao Xutang et al, investigated the thermal deformation in the cocrystal formed by hexabenzo cardamom and tetracyanobenzene through heating the phenomenon of recyclable crystal rupture and self-repair was found in the process of reversible phase transformation of Cocrystal as shown in (Fig. 7 a,b)[97].

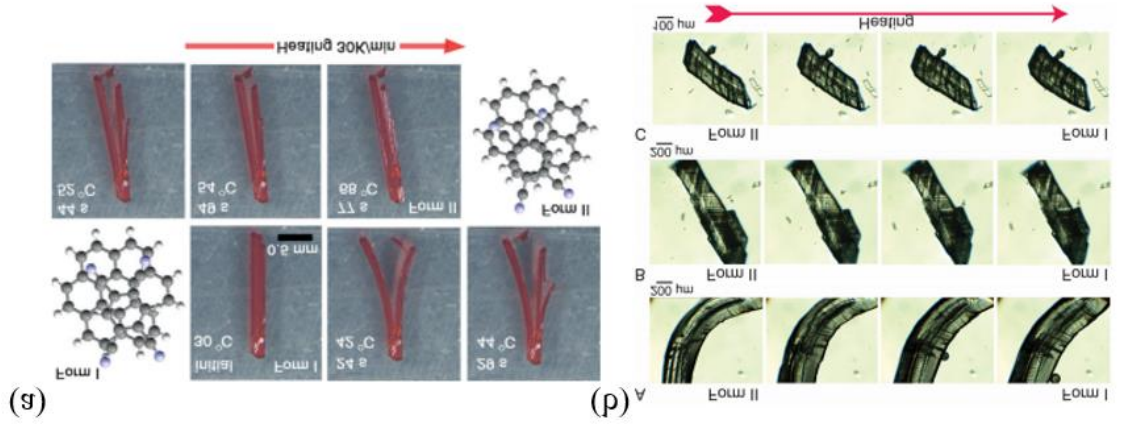


Figure7. (a) (b)The Cocrystal phase transition occurs during heating and causes rupture and self-healing.

The cocrystal exhibits other factors such as liquid crystal material and its magnetic behaviors. In 2004, the British scientist Bruce et al, discovered that some N⋯⋯I the Organic Cocrystal with 1 Bond Exists Liquid Crystal Phase at Certain Temperature, the author believed that intermolecular halogen bonding is a crucial prerequisite for the existence of a liquid crystal phase it means that the halogen bonds are very important in the liquid phase cocrystal[98].

There are some other investigations were reported about the magnetic behavior of organic cocrystals all around the world[99],[100]. A japan scientist T.Hasegawa et al, had been investigated and prepared three kinds of charge transfer complex crystals with alternating columns:(BEDT-TTF)(Me2TCNQ),(BEDT-TTF)(CIMe-TCNQ),(BEDO

TTF)(Cl_2TCNQ), in which (BEDO-TTF)(Cl_2TCNQ) crystal was identified as ionic (with a large degree of ground state charge transfer). The abnormal magnetic properties have been observed on this ionic crystal, and the magnetic susceptibility of the crystal has a sharp decrease near 120K [101]. In 2010. Fumitaka Kagawa et al. studied the charge-transfer TTF-BA Cocrystal and directly measured its field-dependent polarization variation as a one-dimensional organic quantum magnet[102].

4. Application of Low-Dimensional Cocrystal

The organic cocrystal due to the highly important advantages of its crystallinity, well-defined shape, and dimension-dependent optical properties, has been vastly examined in recent years. In recent times the organic low-dimensional co-crystal has been used in the field of laser, optical electronics, organic light emitting transistors, organic light emitting diodes photothermal conversion, and photoelectric conversion, etc. due to new, advanced, and versatile fields the low dimensional cocrystal face various challenges to gain controllable preparation, high charge mobility, and simultaneously, efficient light emission because organic molecules mainly depend on noncovalent interactions and the hydrogen bonding.

4.1 Electrical Properties

4.1.1: Field Effect Transistor

In recent decades, with the advancement of device technology, tens of thousands of organic semiconductor materials have been developed for the research of field-effect transistors. The performance of unipolar charge transport materials, especially hole transport materials, has reached the level of polysilicon, and electronic transport materials have made great progress in recent years. Dodabalapur et al in April 1995 prepared the semiconducting material thiophene oligomer ot-hexathienylene (ox-6T) which has been effectively used in thin film transistors In thin film transistors with a-6T active layers, field-induced conductivity only occurs near the interface plane, while residual conductivity caused by unintentional doping scales with the thickness of the layer[103]. The same author in September 1995 advanced the semiconductor materials that function as n-channel or p-channel devices, which depends on gate bias. The two active ones were a-healthy-phenylene (α-6T) and C_{60}. These two devices mainly depend upon the molecular orbital energy levels and transport activities of α-6T and C_{60}. The synthesized materials were not unique but also used as a building block to form low-cost, low-power complementary integrated circuits[104].

Ran Ding et al synthesized organic one-dimensional as well as two-dimensional co-crystals which demonstrate the development of organic semiconductors and having distinctive molecular shapes and optoelectronic properties. They characterized the novel molecule of C70 having subunits of diindeno[4,3,2,1- fghi:4′,3′,2′,1′-opqr]perylene (1) that is substituted with either triethylsilyl(TES)-ethynyl or 2,4,6-triisopropylphenyl (2) present at meta positions, So by measuring the organic field effect transistor (OFETs), it was demonstrated by authors that molecule 1 shows a hole mobility of 0.31 cm2 V−1 s −1 and the molecules 1-C70 cocrystal exhibits ambipolar transport characteristics with electron and hole mobilities approaching 0.40 and 0.07 cm2 V−1 s −1, respectively. So

this approach of the author leads to the low-dimensional organic cocrystal semiconductor [105].
A method of constructing p-type and n-type double-layer semiconductors in the same device to realize bipolar transmission is proposed and verified. The first Cocrystal with bipolar field-effect properties reported in 2004[106], attracted great attention from the scientific community and led to the discovery of bipolar co-crystal. A boom in the exploration of charge-transfer properties Masatoshi Sakai, Chiba University, Japan (2007) [107]. Wakahara, National Institute of Materials Science, Japan and Bradley, Anthopoulos, et al, imperial Polytechnic Institute UK (2012). Rao and George et al of Jawaharlal Nehru Advanced Scientific Research Centre, India (2010)[23], both the preparation of organic Cocrystal materials and the bipolar charge transport in devices studied.

4.2 Photoelectric Conversion Properties of Organic Co-crystal

The traditional photoelectric conversion technology is based on the mechanism of photogenerated carriers in semiconductors for obtaining solar energy and detecting photons and optical signals. Since it relies on a series of indirect and complex physical processes, it is severely limited in its efficiency and wavelength applicability due to material properties and device structures[108].

Due to their fascinating photophysical properties and potential applications in data storage, bioimaging, and photodynamic therapy, molecular crystal materials with high room-temperature phosphorescence have attracted extensive attention. In this work, the chemical structures of DCPA and AD molecules are presented. A new co-crystal complex [(DCPA) (AD) 2] (DCPA_9, 10-bis (4-carboxyphenyl) anthracene; AD_ Acridine). Figures 8A and 8B show the 1D chain structure of DCPA extended by a C-H$^{\cdots}$O hydrogen bond[109].

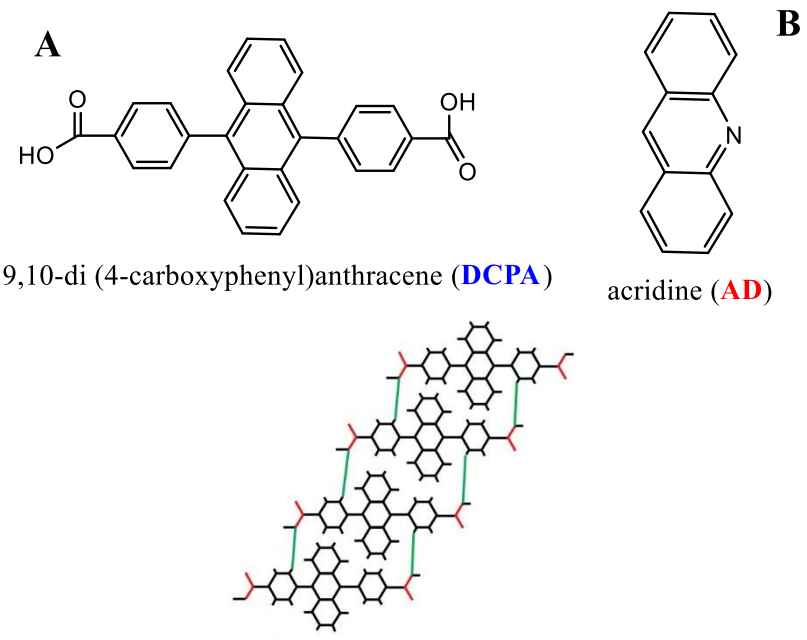


Figure 8. (A) Chemical structures of DCPA and AD molecules in this work. (B) 1D chain-like structure of DCPA extended by C–H···O hydrogen bonds.

In providing a bright future for co-crystals in high-performance electronic devices, 2D co-crystals for broadband photodetectors give them an opportunity for large-scale, integrated, flexible, and practical applications, as well as demonstrating how they can be applied to electronic devices. In addition to strong cyan fluorescence (58.20 nanoseconds), organic photoresponsive materials also exhibit direct white phosphorescence (325 seconds) as determined by photoluminescence measurements[110].

A transient current density time curve shows that the [(DCPA) (AD) 2] co-crystal exhibits a strong photoelectric response when exposed to simulated light. A large space interval between HOMO and LUMO is required for improved charge transfer of donors-acceptors, while a small EST is required. Several single-component organic molecules with distorted spatial conformations of donor receptors have been efficient RTP materials in the past. In addition, the mixed co-crystal derived from electron-rich donor (D) and electron-deficient acceptor (A) molecules shows a wide range of electronic, optical, and magnetic properties[109].

It was prepared using chloroform crystallization, vacuum co-sublimation, and pure grinding to create a co-crystal of p-benzoquinone (BQ) and one of the four mono-substituted p-halophenols. It is possible to prepare co-crystals with 1:1 or 2:1 donor receptor ratios from all four BQ halophenol combinations[111]. These crystals can be used in photodetectors in the future. They are bonded by powerful hydrogen bonds[109]. Many factors affect the photovoltaic behavior of organic materials, including the donor-acceptor contact mode, which regulates the generation process of photocurrents, including exciton diffusion, dissociation, and charge transport. At room temperature, Insert shows solid-state samples under UV (365 nm) light radiation[112]. steady state and

transient photoluminescence (PL) spectra, as well as time-resolved PL decay curves, were recorded for [(DCPA) (AD) 2] co-crystal, pure DCPA, and AD solids which are shown in Figure 9.

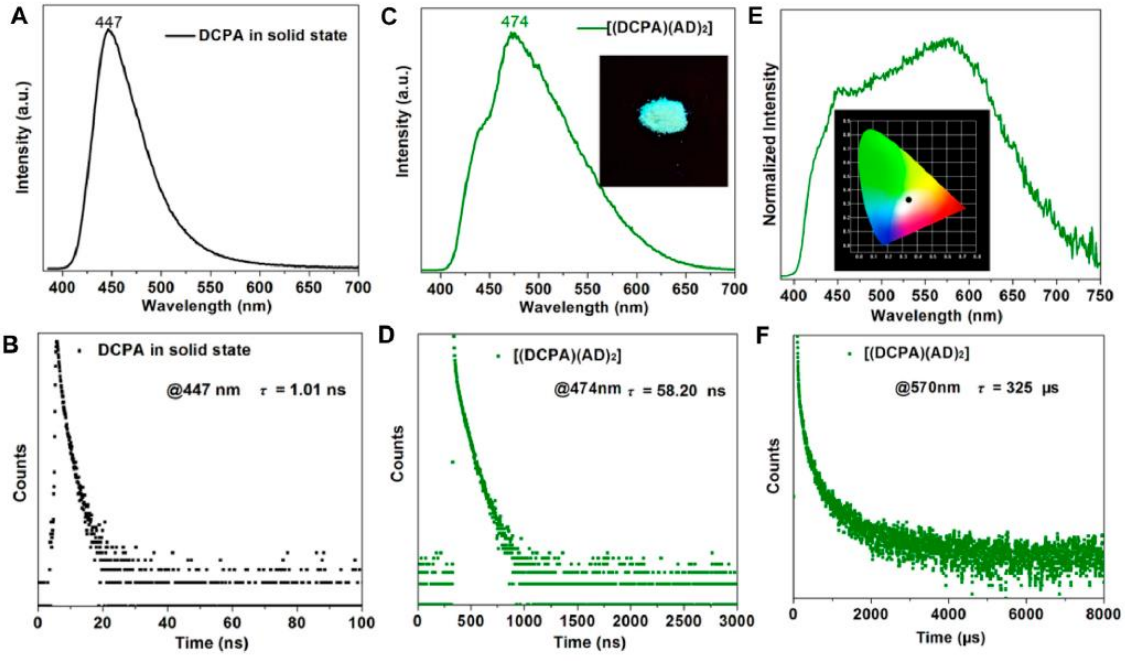


Figure 9. Fluorescence spectra (A) decay curve (B) of DCPA in solid state (λex _ 329 nm). Fluorescence spectra (C) decay curve (D) of [(DCPA) (AD)2] co-crystal in solid state (λex _ 365 nm). Insert shows solid-state samples under UV (365 nm) light radiation. Phosphorescence spectra (E) decay curve (F) of [(DCPA) (AD)2] co-crystal in solid state (λex _ 365 nm).

By combining the characteristics of its components, including white light emission, ambipolar charge transfer, nonlinear optics, and ferroelectricity, can achieve white light emission. Co-crystal engineering is a new approach to creating multifunctional materials, and the CIE-1931 chromaticity diagram (0.33-0.34) shows [(DCPA) (AD) 2] co-crystal with white phosphorescent emission. The measurements were taken under environmental conditions[109].

Wenping Hu et al. designed and grew two typical luminescent organic co-crystals, including pyrene-octafluoronaphthalene (pyrene-OFN) and pyrene-1,2,4,5-tetracyanobenzene (pyrene-TCNB), through Strategies for the development of simple supramolecular assembly. Co-crystals exhibit different optical properties due to their different modes of intermolecular interaction; namely, arene-perfluoroarene (AP) and charge-transfer (CT) interactions. Unexpectedly, the pyrene-TCNB system with strong CT interaction was incorporated into the pyrene-OFN host as a powerful guest to produce white light emission (WLE). The current competitive intermolecular interaction strategy can be applied to fabricate more complex organic light-harvesting systems [119].

4.3. Photothermal Conversion Properties of Organic Co-crystals

Several organic photothermal materials (such as organic co-crystals) have also attracted considerable interest in basic research and practical applications because they combine a photothermal conversion effect with high flexibility and efficiency. The solar thermal conversion process converts sunlight into heat by collecting and abusing it through a system. In terms of the photothermal effect, the infrared band in the solar spectrum is responsible for directly generating heat, while most light cannot. The photon energy interacts with the crystal lattice when the light is irradiated on the light-absorbing material, increasing vibration and temperature which needs a wide spectral absorption range [113]. Dong Zhao et al, suggested an excellent solution by designing an organic charge transfer cocrystal having an energy gap of 0.33 eV and photothermal conversion efficiency of 69.3% which was a practically simple self-assembly process and introduced into polyurethane to form a large area of photothermal nanofiber films by electrospinning technology. To control CT complexes' energetics and properties, D/A parent compounds must be selected. Additionally, the temperature of the photothermal nanofiber films prepared under laser radiation at 0.183W/cm^2 is rapidly increased to 52 °C, indicating a PCE of 53.7%. As a result of this work, large-area photothermal films can be manufactured, as well as photothermal imaging can be developed. Near-infrared photothermal conversion is of great interest in many sectors[114].

Due to its superior performance, CQW has great potential for photoelectric applications, including solar energy collection, lasers, and light-emitting diodes (LEDs)[115]. The Figure shows the structures of a self-assembled organic co-crystal (N, N, N, N-tetramethyl-p-phenylenediamine) (TMPD) and pyromellitic dianhydride (PMDA) which is shown in Fig 10A. It is NIR photothermal conversion efficiency is high (87.2%) with strong absorption in the NIR range and broad absorption (200-1500nm). As a result of the small HOMO-LUMO gap of the donor-acceptor pair in the co-crystal, it can absorb in the NIR region with greater efficiency. As a result, Fig. 10B presents electron spin resonance (ESR) spectra of TMPD, PMDA, and TMPD-PMDA co-crystals of pyromellitic dianhydride (PMDA). Due to strong intermolecular interactions, the co-crystal has dense electronic energy levels, resulting in vibrational cooling (94.4%) and an internal conversion process with ultrafast excited state relaxation (0.12 ps), resulting in high photothermal conversion efficiency in the NIR. In addition to providing excellent optical and thermal performance, selective absorber surfaces are excellent candidates for NIR photothermal therapy[116]. Sun Lingjie et al have given a progress report on developing effective design principles for current high-performance functional materials they also explained the photothermal (PT) materials that can convert infrared light into heat energy in photothermal therapy, photothermal electrical equipment, photothermal/photoacoustic imaging, and shape memory devices [117].

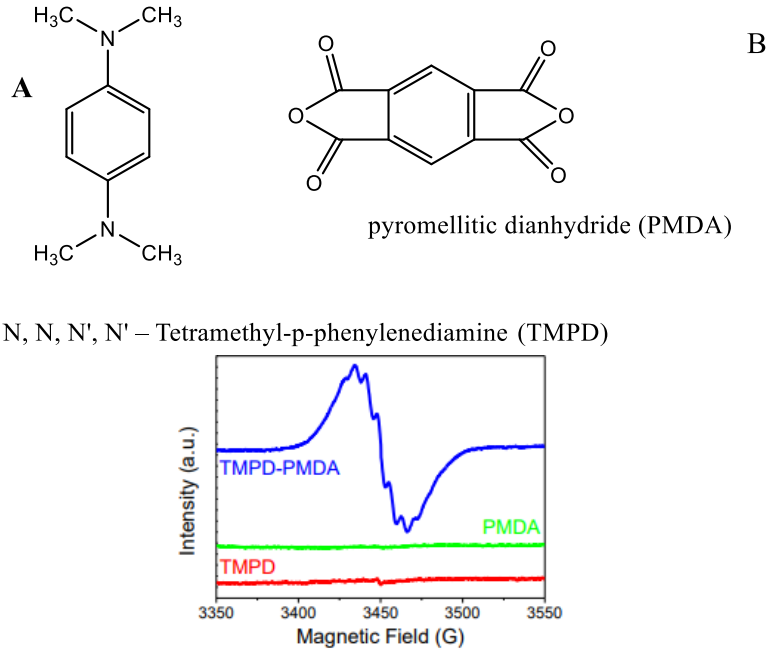


Figure 10. (A) the structure is N, N, N', N' – Tetramethyl-p-phenylenediamine (TMPD) and (PTMDA), (B) pyromellitic dianhydride (PMDA) ESR spectra of TMPD, PMDA, and TMPD-PMDA co-crystal.

Yang et al manufactured 3D-printed photoresponsive shape memory devices by combining FDM printing technology with PU and CB-based photoresponsive shape memory composites. In the 3D printing process of photoresponsive devices, FDM is applied to avoid the influence of UV illumination. The addition of CB and efficient photothermal conversion enable 3D printing shape memory devices to have an excellent light response. Interestingly, natural sunlight can also trigger the shape memory behavior of 3D printing devices. The development of this simple printing strategy will give enormous opportunities for the design and manufacturing of personalized stimulus-response devices, which can be widely applied in attractive and highly personalized biomimetic intelligent devices and soft robots[118].

4.3.1. Photothermal Imaging

The authors arranged the co-crystal powder into three letters, S, T, and U, then irradiated it with an 808nm laser. Infrared thermal imaging was then used to observe the powder. Due to the excellent ability of TMPD-PMDA co-crystals to convert PT, when they are irradiated, letters are immediately visible on the thermal image (Fig. 11 A)[116]. Hu Wengping et al. explained the excited state of DTC tends to undergo rapid nonradiative decay, while the radiative transition process is suppressed by efficient fluorescence quenching in the solid state. Due to these properties, DTC exhibits high-performance PT conversion characteristics and high cycle stability. At the same time, the potential was encouraging nondestructive PT imaging with real-time and time-resolved responses (Fig. 11 B)[38]

Kim et al, supposed that Photothermal cocrystal materials convert and absorb electromagnetic radiation energy into heat energy through the non-radiative exothermic reaction of materials. The authors give in detail the photothermal effect of the materials which have many applications in near-infrared regions such as PT ablation, spacecraft thermal controls, solar cells, and non-invasive selective harvesting of stem cells. However, it has the advantages of the infrared region, so it was applied to dark field observation applications such as safety cameras, night vision sensors, and optical attenuators[119].

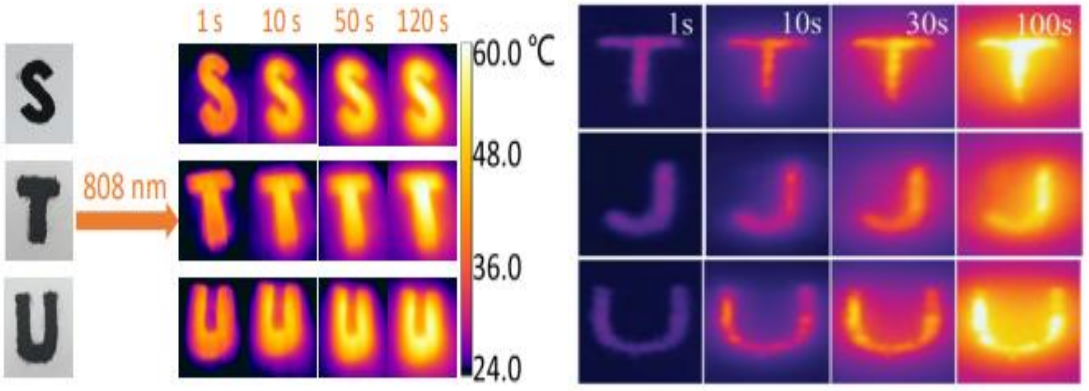


Figure 11. Demonstration of photothermal imaging utilizing TMPD-PMDA co-crystal under 808 nm laser.

5. Conclusion

In conclusion, co-crystals are an aggregate of molecules, with at least two molecules in their structure. Additionally, there are two main types of co-crystals, organic and inorganic. Researchers were soon interested in co-crystals because of the vastness of the co-crystals performance field and the possibility of using them for numerous purposes. In this review, we have briefly discussed the organic low-dimension co-crystals that have been extensively researched in electronics, electroluminescent devices, photothermal, photoelectric conversion, and many other areas. The organic low-dimension co-crystal concentrates the electron-transporting acceptor material and the hole-transporting donor material in a single crystal structure and integrates the electron transport channel and the whole transport channel through simple non-covalent interactions, which is to achieve bipolarity and effective strategy for transmission. However, due to the unclear organic low-dimension co-crystal mechanism reports on their structure control and morphology changes are limited however, coupled with the unpredictable crystal structure, there are only a handful of co-crystal materials that can achieve ambipolar transport. Therefore, how to rationally select receptor molecules, realize efficient recognition between different components, and obtain a molecular stacking structure that is conducive to charge transport is the primary problem in promoting the application of low-dimension organic co-crystals in bipolar charge transport. The low-dimension co-crystal can be prepared in three different ways, including liquid phase methods, gas phase methods, and solid phase methods. By applying these methods, organic Cocrystals of various sizes can be produced, including nanocrystals, small crystals, and microcrystals. In this review, we have briefly discussed different effects on low-dimension organic co-crystals such as, they are

stimulated by external stimuli (heating, light radiation, and external force) In addition we have discussed binary complexes which can be packed in two ways: one is segregated stacking, where the same molecule like a donor or acceptor is packed into its column, and the other is mixed packing, where different molecules pack in alternative ways, modeled. The intermolecular interaction and structure of co-crystals determine their physical and chemical properties, including their photoelectric and thermal conversion capabilities. It is, therefore, necessary to conduct an immense and vastly extended systematic study of organic low-dimension co-crystal control structure and morphology-changing behavior by different stimulations.

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Conflict of interest
The authors declare that they have no conflict of interest.

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