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Piezoelectric materials for pollutants degradation: State-of-the-art accomplishments and prospects

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ABSTRACT

Piezoelectric catalysis, a new catalytic method, is widely used in the field of environmental sanitation, including waste water treatment and dye degradation. However, in the face of the growing environmental pollution problem, the efficiency of piezoelectric catalysis is still hampered by the stress variation in the natural environment. Therefore, it is particularly important to improve the catalytic efficiency of piezoelectric materials. We divide piezoelectric materials into two categories: inorganic piezoelectric materials and organic piezoelectric materials. Then the mainstream inorganic piezoelectric materials are divided into four subcategories, namely: (1) MTiO_3 ($M = \text{Ba}, \text{Sr}$), (2) bi-class catalytic materials, (3) MoX_2 ($X = \text{S}, \text{Se}$), and (4) ZnO piezoelectric materials. The mainstream organic piezoelectric materials are divided into PVDF and $\text{g-C}_3\text{N}_4$ materials. At the same time, the above materials are summarized to explain the excellent performance of materials from the perspective of structure and piezoelectric principle. In addition, we summarized the modification methods that can be applied to piezoelectric materials: (1) Morphology methods, (2) composites with heterojunctions, and (3) surface modification. Finally, we summarized the prospects of piezoelectric materials in the field of environment and water treatment.

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1. Introduction

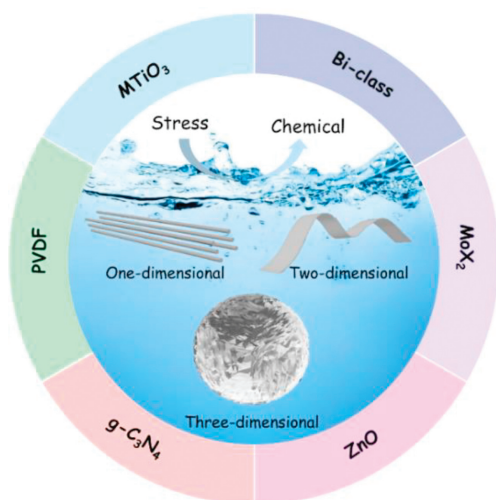
With the rapid development of industrialization, environmental water contamination has become increasingly unavoidable [1]. Water bodies have the ability to self-purify, which allows them to reduce or even reverse the negative impacts of introduced contaminants. And as the quality of natural water declines, self-purification plays an increasingly important role [2,3]. Water in natural ecosystems can purify itself using a method that includes physical, chemical, or biological elements [2,4]. In addition, water bodies contain mechanical energy during their flow, including the ebb and flow of the tide. Due to hydraulic cavitation, waterfall scouring can also be used as potential power to cure water pollution. For example, waterfalls flow from high to low, creating cavitation and piezoelectric effects as the water rushes by. The energy is converted from potential energy mostly to mechanical energy and partly to chemical energy.

Therefore, worldwide researchers want to simulate and enhance the purification process of natural water bodies through laboratory approaches or means to achieve the effect of degrading pollutants and at the same time broaden the ideas of water pollution treatment [5,6].

There are two main aspects to study the hydraulic cavitation effects in the laboratory: (1) creating experimental conditions for hydraulic cavitation by adjusting the way stress is applied, such as water flow, ultrasound, designing cavitation devices; (2) studying piezoelectric materials to harvest efficient mechanical energy for hydraulic cavitation. In general, when driven by mechanical pressure or temperature fluctuation, piezoelectric materials can produce an internal electric field to transfer the free charge carriers in the opposing direction [7,8]. Dissolved oxygen in water and water is activated as reactive oxide species (ROS) at both ends of the internal electric field of the piezoelectric material to participate in the redox reactions in water. Piezoelectric materials hold great promise in the fields of catalysis, and are often used for sewage treatment under the condition of ultrasonic cavitation

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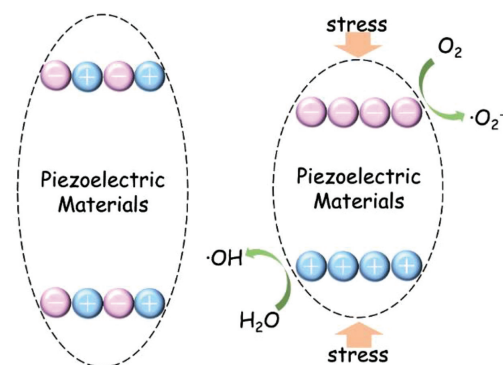


Scheme 1. Classifications of the more commonly studied piezoelectric materials.

[9–14]. At present, the more widely researched piezoelectric materials are: BaTiO₃ [15–23], Na_{0.5}Bi_{0.5}TiO₃ [1,24–26], ZnO [27–38,42], BiOX (X = Cl, Br, I) [39–41,43–45], BiFeO₃ [46–48], Bi₂S₃ [49], BiOI₃ [50], MoS₂ [51–60], PVDF [34,35,61–64], and g-C₃N₄ [59,64–68] (Scheme 1). These piezoelectric materials can be categorized into five classes, which are MTiO₃ (M = Ba, Sr), Bi-class catalytic materials, MoX₂ (X = S, Se), ZnO catalytic materials, and organic piezoelectric material. In addition, the key role of solar irradiation in enhancing the piezoelectric catalysis based on piezoelectric-photoelectron effect have been widely concerned [7]. Subsequently, the generation of ROS from the material class and the mechanisms for degradation of organic pollutants (such as dye, antibiotics, and industrial effluent) were summarized (Table 1). At end, piezoelectric catalysis enhanced self-purification of water bodies is proposed and summarized with an outlook.

2. Piezoelectric principle

The features of piezoelectric materials are occurring polarize or change the polarization state, under the condition that the force field or electric field has changed. Piezoelectric materials act as a bridge between mechanical energy and chemical energy, converting stress changes into chemical energy [69]. It is possible to employ the chemical energy to breakdown contaminants in natural water bodies. The dielectric constant of a piezoelectric material is its most significant physical property. In general, the greater the dielectric constant of a material, the greater the intensity of polarization produced under an electric or force field of the same strength [70]. The mechanism of dielectric polarization can be divided into three types: electron displacement polarization, ion displacement polarization, and orientation polarization [71]. Because the positive and negative charge centers of ions or atoms wouldn't coincide, electron displacement polarization results in the majority of the usual piezoelectric materials with perovskite. The piezoelectric effect, which occurs when a piezoelectric crystal is deformed by an external force, is the most significant phenomena for piezoelectric materials [72]. It refers to the charge buildup that occurs on a material's surface and is linearly proportional to the external force. From crystallographic research, it is possible to anticipate which crystals will have the piezoelectric property. Crystals without a center of symmetry exhibit piezoelectricity. Pressure applied to a dielectric material causes the crystal lattice to distort and separate the centers of gravity of oppositely charged species, which results in the creation of a dipole moment in each molecule (Scheme 2) [73].



Scheme 2. Piezoelectric catalysis mechanism.

These polarized charges will generate active species to take part in the redox reactions in natural water during the process of piezoelectric catalysis. Generally, the stress is originated from the fluctuations in water flow and agitation. In laboratory scale, the most common source of piezoelectric force is ultrasound. Ultrasound causes the medium to vibrate and when the ultrasound reaches a certain intensity, cavitation occurs. Cavitation effect is the process of a bubble developing, expanding, and deflating. When the bubble collapse, the water bodies generates a high local energy density [74]. Cavitation causes the local environment to reach extremely high temperatures and pressures (100–5000 atmospheres of pressure and 500–15,000K), yet the broader environment is kept at ambient air conditions [75,76]. This enables the effective execution under ambient conditions of the various physical processes or chemical reactions that require stringent conditions [77,78]. Ultrasonic cavitation usually goes through three processes: (i) the formation of cavitation bubbles, (ii) growth of cavitation bubbles, and (iii) the collapse of cavitation bubbles. The cavity formed in the liquid during ultrasonic will produce fixed point high temperature and high-pressure during collapse blasting. The pressure generated by the cavitation bubble during collapse exerts a force on the piezoelectric material, resulting in deformation of the material and producing piezoelectric potential energy, and the resulting high fixed-point temperature can be involved in the activation of the applied oxide species, such as PMS, PDS [79,80], H₂O₂, CH₃COOOH. Ultrasonic cavitation is being used in a variety of sectors for things like the forced breakdown of polymer materials, the rupturing of chemical bonds, the production of free radicals, and more. When ultrasonics has a higher power, more cavitation bubbles are produced in the liquid, accompanied by the generation of ROS, such as ·OH. The power of ultrasonics is also an important indicator to determine the number and size of cavitation bubbles. Specifically, lower ultrasound frequencies produce larger and smaller cavitation bubbles, while higher ultrasound frequencies produce more but smaller cavitation bubbles [81]. Currently, piezoelectric catalysis uses ultrasonic cavitation primarily, and the frequency range is typically 20–40 kHz.

3. Inorganic piezoelectric materials for pollutants removal

3.1. MTiO₃ (M = Ba, Sr)

In recent years, elemental Ti is the star material in both photocatalysis and piezoelectric catalysis, playing an important role in catalytic materials. In the field of piezoelectric catalysis, BaTiO₃ [76,82,83], as a classical material, has a long research history and a broad research prospect. Among the common titanium-containing piezoelectric materials, most of them are based on BaTiO₃ and are modified on this basis. BaTiO₃ and SrTiO₃ are two of the most popular piezoelectric materials which contain Ti elements. As a

Table 1

Summary on organic pollutants degradation by piezoelectric materials.

	Catalyst	Contaminant	Light source	Ultrasonic vibration	Performance	Time	ROS	Ref.
Ti	BaTiO ₃ NWs	MO (5 mg/L)	4.2 W, UV-LED point lamp	180 W, 40 kHz	98.17%	80 min	[•] OH, [•] O ₂ ⁻	[21]
	BaTiO ₃	RhB (5 mg/L)	–	400 W, 40 kHz	94%	120 min	[•] OH, [•] O ₂ ⁻	[23]
		Ciprofloxacin (10 mg/L)	300 W Xe lamp ($\lambda > 420$ nm)	210 W, 40 kHz	50.20%	90 min	[•] OH, [•] O ₂ ⁻	[22]
Bi	BaTiO ₃ /La ₂ Ti ₂ O ₇	MO (10 mg/L)	200 W Xe lamp	100 W, 40 kHz	90%	45 min	[•] O ₂ ⁻ , h ⁺	[20]
	BaTiO ₃ /CuO	RhB (1 × 10 ⁻⁵ mol/L)	15 W visible light lamps	110 W, 40 kHz	43.20%	30 min	[•] OH, [•] O ₂ ⁻	[49]
	BiOX/BaTiO ₃ (X = Cl, Br)	RhB, MB (10 mg/L)	300 W Xe light lamps ($\lambda > 380$ nm)	100 W, 40 kHz	100%	3 min	[•] OH, [•] O ₂ ⁻ , ¹ O ₂	[39]
	BiFeO ₃ @CdS	BPA (10 mg/L)	–	300 W, 40 kHz	99.7%	60 min	[•] OH, [•] O ₂ ⁻	[47]
	Na _{0.5} Bi _{0.5} TiO ₃	RhB, MO (5 mg/L)	–	80 W, 40 kHz	100%, 92.8%	80 min	[•] OH, [•] O ₂ ⁻ , h ⁺	[1]
	BiOI ₃	RhB (10 mg/L)	300 W Xe light lamps	120 W, 40 kHz	98.6%	60 min	[•] OH, h ⁺	[50]
Mo	MoSe ₂ NFs	RhB (10 mg/L)	–	250 W, 40 kHz	100%	2 min	H ₂ O ₂ , [•] OH, [•] O ₂ ⁻	[60]
	MoS ₂ NFs	RhB (10 mg/L)	–	250 W, 40 kHz	100%	5 min	[•] OH, [•] O ₂ ⁻ , H ₂ O ₂	[54]
	MoS ₂ /C	RhB (10 mg/L)	–	100 W, 20 kHz	100%	15 min	[•] OH	[56]
	MoS ₂ /carbon fiber	RhB (10 mg/L)	–	1.67 mL/min, 10 L/min	100%, 100%	3 min, 40 min	[•] OH	[57]
	Co-MoS ₂	MB (20 mg/L)	–	100 W, 40 kHz	99.6%	60 s	[•] OH, [•] O ₂ ⁻	[59]
ZnO	Ultrathin-ZnO/Al ₂ O ₃	IMD (20 mg/L)	–	100 W, 40 kHz	96%	15 min	[•] OH, [•] O ₂ ⁻	[32]
	A-ZnO, H-ZnO, V-ZnO	MO (50 mg/L)	–	30 W, 30 kHz (40 kHz, 50 kHz, 60 kHz)	100.00%	120 min	[•] OH, [•] O ₂ ⁻ , h ⁺	[27]
C ₃ N ₄	2D g-C ₃ N ₄	2,4-DCP (20 mg/L)	–	120 W, 40 kHz	100%	150 min	[•] OH, [•] O ₂ ⁻	[59]
	g-C ₃ N ₄ /PDI-g-C ₃ N ₄	2,4-DCP, 2,5-DCP, 2,6-DCP (20 mg/L)	300 W Xe lamp ($\lambda > 420$ nm)	120 W, 40 kHz	89.7%, 73.6%, 62.6%	60 min	[•] OH, [•] O ₂ ⁻ , ¹ O ₂ , h ⁺	[66]
	Bi ₂ WO ₆ /g-C ₃ N ₄ /ZnO	RhB, MO (5 mg/L)	100 W incandescent lamp ($\lambda > 420$ nm)	80 W, 120 kHz	100%, 100%	20 min	[•] OH, [•] O ₂ ⁻ , H ₂ O ₂ , h ⁺	[68]
PVDF	Fe ₂ O ₃ /PVDF-HFP porous film	Atrazine (10 mg/L)	–	200 W, 40 kHz	77.30%	660 min	[•] OH, [•] O ₂ ⁻	[61]
	HPVDF/BTO-Ovs	TC (50 mg/L)	–	300 rpm	53.70%	60 min	[•] OH, [•] O ₂ ⁻	[63]

"–" means it is not mentioned in reference.

classical piezoelectric material, BaTiO₃ has high dielectric constant and low dielectric loss. The BaTiO₃ formed at 1460–130 °C exhibits cubic chalcogenide type structure, the crystal structure is highly symmetrical as there are no dipole moments in the material and hence, there is no piezoelectricity. As the temperature drops, the symmetry of the crystal begins to decline. At 130 °C, BaTiO₃ underwent a ferroelectric phase transition. At 130–5 °C, BaTiO₃ is tetragonal system point group, which has significant ferroelectricity and spontaneous polarization intensity along the direction of (001). The spontaneous polarization of BaTiO₃ mainly comes from electronic displacement polarization and ion displacement polarization. In addition to BaTiO₃ related piezoelectric materials, SrTiO₃ also has piezoelectric properties, ion-shift polarization and electron-shift polarization lead to a large spontaneous polarization inside the SrTiO₃ crystal [84].

Different synthesis methods lead to different morphologies of BaTiO₃ materials, namely BaTiO₃ nanowires, BaTiO₃ nanoparticles and dendritic BaTiO₃. The degradation of Cu-EDTA under the combined effect of ultrasound and catalysis was stronger than that under single conditions (Fig. 1A). Based on the simulations from COMSOL Multiphysics, the dendritic and linear BaTiO₃ exhibit better piezoelectric potential (Fig. 1B) [85]. SrTiO₃–1, SrTiO₃–2, and SrTiO₃–3 nanoparticles with different exposed crystalline faces and co-exposed faces were synthesized by design. SrTiO₃–1 only ex-

hibits the (001) face, while SrTiO₃–2 and SrTiO₃–3 expose both the (001) and (110) faces. The authors found that SrTiO₃–2 degraded at a higher rate than SrTiO₃–1 and SrTiO₃–3 under the same conditions, at 80% after 3 h of sonication, which was indicative of the material's piezoelectric properties (Figs. 1C and D) [86]. According to earlier reports, surface reconstruction can generate stronger polarization when subjected to ultrasonic vibration, which is advantageous for the generation of ROS. By comparing the amount and type of ROS generation, it was found that the SrTiO₃ (001) surface produces more [•]OH and the (110) surface produces more [•]O₂⁻. The (110) and (001) surfaces form a "surface heterojunction" with different ratio affecting the piezoelectric catalytic performance of SrTiO₃. At present, titanium-containing materials have become a bridge between piezoelectric catalysis and photocatalysis. Due to the synergistic interaction of piezoelectricity and light, the photogenerated electrons and holes produced by light are separated by the force of the piezoelectric potential, which lowers the rate of complexation between the two. As a result, piezoelectric materials have a broader future in the environmental field.

3.2. Bi-class catalytic materials

Crystals containing bismuth element have been of great interest to the materials community, such as BiFeO₃ catalyst, BiOX (X = Cl, Br, I), Bi₂S₃, Bi₂WO₆, Bi₂MoO₆, and other cutting-edge materials.

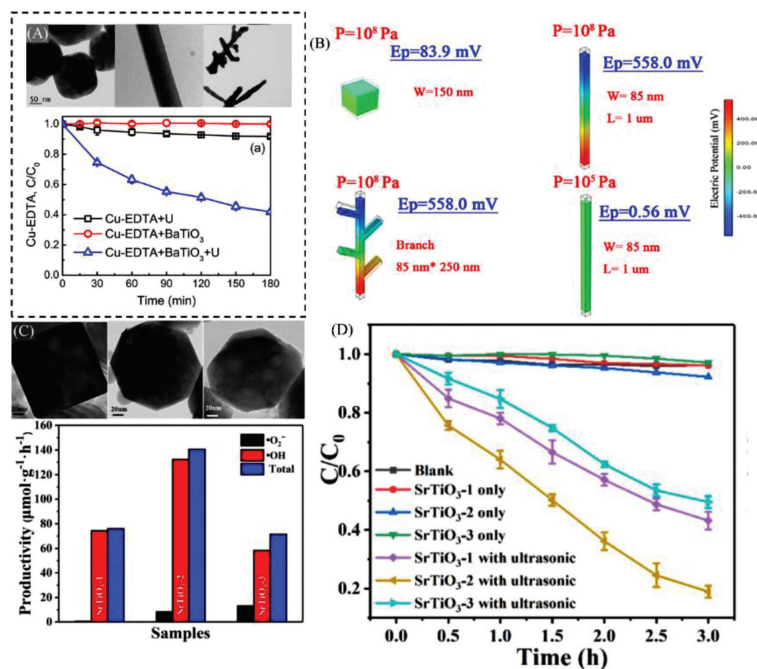


Fig. 1. (A) Piezo-catalytic degradation of Cu-EDTA by BaTiO₃ nanowires with respect to reaction time as observed in TEM images of BaTiO₃ crystallites with various morphologies under various conditions. (B) To simulate the piezoelectric potential distribution of BaTiO₃ nanocrystals with various morphologies, use COMSOL Multiphysics. Arborization under cavitation pressure of $P = 10^8$ Pa, nanowire under cavitation pressure of $P = 10^8$ Pa, and nanoparticle under cavitation pressure of $P = 10^5$ Pa. (A, B) Copied with permission [85]. Copyright 2022, American Chemical Society. (C) SrTiO₃-1, SrTiO₃-2, and SrTiO₃-3 TEM images showing active radical generation across serial SrTiO₃ samples. (D) SrTiO₃-1, SrTiO₃-2, SrTiO₃-3 and creation of active radicals over successive samples of SrTiO₃. (D) Ultrasonic vibration and piezoelectric catalytic degradation of RhB in the presence of sequential SrTiO₃ samples. (C, D) Copied with permission [86]. Copyright 2022, Elsevier.

The lone pair of electrons in the Bi³⁺ ion leads to a general asymmetry in the ionic crystals associated with Bi³⁺. For piezoelectric materials, the asymmetric crystal structure is prone to distortion when stress changes are felt, which leads to a shift in the position of the central ion, a shift in the charge density, and the generation of dipoles inside the crystal with piezoelectricity.

Bi₂S₃ shows the orthorhombic structure's two [BiS₇] clusters that make up the unit cell representation. As can be observed, both clusters exhibit severe structural abnormalities and asymmetry. When there are no outside disturbances and no visible light, Bi₂S₃ showed very low oxidation activity. Under the action of ultrasound, the separation of electron-hole pairs is promoted. As a result, ROS is formed on the surface of the material, which is used to degrade organic pollutants (Figs. 2A and B) [49]. In addition, BiOX (X = Cl, Br, I) not only possesses good photocatalytic properties, but also can be applied to piezoelectric catalysis because BiOX has a layered structure characterized by alternating Bi₂O₂ slabs and halogen atomic bilayers [87]. Nanoclusters or nanoflowers are usually assembled from nanosheets, and in piezoelectric catalysis, each nanosheet can act as a positive and negative electrode for the piezoelectric potential. Theoretical calculations show that the conduction band (CB) of bismuth-based compounds is mainly composed of Bi 6p orbitals, and the CB potential is dependent on the content of Bi in the materials [21]. Meanwhile, the valence band (VB) of bismuth-based photocatalysts is made up from the hybrid orbitals of O 2p and well-dispersed Bi 6s. The Bi 6s orbital can accelerate the mobility of photogenerated charge carriers and also narrow the band gap [88–90]. BiOI, a member of the BiOX series of compounds, has the best in-plane piezoelectric characteristic, with a value of 34.36 pm/V, which is significantly greater than the value of MoS₂ (3.73 pm/V), which was previously reported [91]. The in-plane piezoelectric coefficient of any bismuth oxyhalide is higher than that of BiXY when compared with the same series of BiXY (X = S, Se, Te and Y = F, Cl, Br, I) [91].

Similar to Bi₂S₃ and BiOX, the spontaneous polarization of Bi₂WO₆ and BiFeO₃ also results from the relative displacement of Bi³⁺. Octahedral (WO₄)²⁻ and (Bi₂O₂)²⁺ slabs alternate to generate the layered structure of bismuth tungstate (Bi₂WO₆). Bi₂WO₆'s distinctive sandwich structure allows for the induction of an internal electric field between layers, which is advantageous for separating electrons from holes [92]. It possesses beneficial physical and chemical characteristics, including piezoelectricity and catalysis, both of which have lately undergone substantial research [93]. It frequently only has layers that are a few microns thick and typically displays a 3D nano-flower design (Fig. 2C). Under ultrasound drive, BWO was reported to degrade 76% of RhB within six hours, with a much higher performance than the other two control experiments performed using a single condition (only ultrasound or only catalyst) [80]. The free radical capture experiments showed that the ROS contributing the most to the degradation process were •OH and •O₂⁻ (Figs. 2D and E). Another important Bi-class material, BiFeO₃ (BFO) has a rhombohedrally distorted perovskite structure with space group R3c. Its ferroelectric order originates from the stereochemical activity of the Bi³⁺ lone electron pair in BiFeO₃ (BFO), leading to both anti-ferromagnetism and weak ferromagnetism [94]. The center and diagonal of the octahedron of FeO₆ are Fe³⁺ and Bi³⁺, respectively. Owing to the hybridization between the lone pair electrons in the Bi³⁺ 6s orbital and O²⁻, there is a deviation of Bi³⁺ in the octahedral center. Thus, ferroelectric polarization appears in the direction of (111) [95]. Two different morphologies of BFO were prepared by hydrothermal approach, which are square nanosheets (NSs) and nanowires (NWs). By comparing the performance of piezoelectricity, light feeding and photopiezoelectricity synergy, it can be found that the degradation capability of BFO for RhB dye degradation under photo-piezoelectricity synergy is stronger than that of single condition, and the degradation effect of NWs is stronger than that of NSs. The piezoelectric field inside the BFO, which encourages charge carrier separation, is

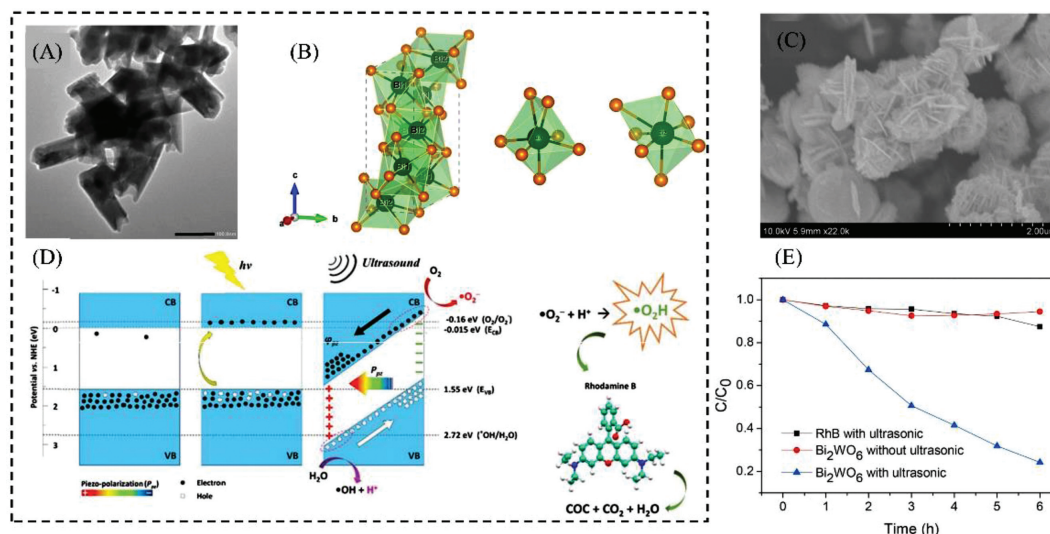


Fig. 2. (A) TEM image of Bi₂S₃. (B) Unit cell and composing clusters of Bi₂S₃ orthorhombic structure. (D) Bi₂S₃'s energy band diagram, shown schematically, under the influence of ultrasound, visible light, and external disturbance. (A, B, D) Copied with permission [49]. Copyright 2021, Elsevier. (C) FESEM image of Bi₂WO₆. (E) Bi₂WO₆ degrades RhB under different conditions. (C, E) Copied with permission [93]. Copyright 2020, Elsevier.

the source of the rapid deterioration under mechanical vibrations. The free charges and cavities lead to the generation of ROS such as $\cdot\text{OH}$ and $\cdot\text{O}_2^-$ in the water column, which subsequently break down the dye molecules into smaller molecules [96].

3.3. MoX₂ (X=S, Se)

TMDs [75,97,98], whose generalized formula is MX₂ (M = transition metal (Ti, V, Nb, Mo, Pd, Pt), X = chalcogen (S, Se, Te)), have a wide range of materials and their electronic properties can be semiconducting, metallic, and superconducting [13]. Mo (+4) and S (−2) are structured to a sandwich structure in a single layer of MoS₂ films by covalent bonds in the order S-Mo-S, with the sandwich layers interacting via relatively weak van der Waals forces. Each layer typically has a thickness of around 0.65 nm. It is discovered that monolayer MoS₂ with trigonal prismatic polypeptide is semiconducting (referred to as 2H). Two-dimensional materials are more easily deformed, which leads to distorted deformation of the internal structure of the material, resulting in piezoelectric potential. The cavitation bubbles generated during ultrasound, the impact force generated during bursting forces the lamellar material to deform, thus generating a piezoelectric potential. The size of the deformation that is occurring in the material determines the magnitude of the piezoelectric potential energy; the greater the piezoelectric potential, the greater the degree of energy band bending, the more potent the active species produced, and the better the degradation of pollutants. MoX₂ (X=S, Se) are typical transition metal two-dimensional materials, each layer consists of a plane of Mo atoms sandwiched between two planes of S or Se atoms [97,98]. They are similar in thickness to graphene and has semiconductor properties with high electron transfer rate. Therefore, MoX₂ are suggested as a piezoelectric material due to the layered structures and curled edge structure, which results in stronger piezoelectric activity (Fig. 3A). It was reported that MoS₂ nanosheets can degrade RhB within 60 s with a degradation efficiency of 93% under ultrasound condition (Fig. 3B) [54]. In addition, few layer MoSe₂ nanoflowers also exhibited strong piezoelectric properties and degraded 90% of the RhB within 30 s in the presence of ultrasound (Figs. 3C and D) [60].

3.4. ZnO

The electrons in the valence band of ZnO, an II-VI compound semiconductor whose ionicity is on the cusp of covalent and ionic semiconductor, can accept the energy transition in UV light [99–103]. The crystal structure of ZnO is made up of a Zn atom and four O ligands to form a tetrahedral structure as well as a non-centrosymmetric structure. Due to the displacement of O^{2−} anions and Zn²⁺ cations caused by the external tension placed on the unit cell, ZnO is regarded as a typical piezoelectric material [32,103,104]. Therefore, ZnO is widely used in catalytic applications including photocatalysis, piezoelectric catalysis, and photopiezoelectric catalysis [105–107]. The diversity of ZnO nanomaterials is greatly dependent on the growth mechanism, growth methods, and synthesis conditions. Due to the relatively simple composition of ZnO, the current research progress is mainly focused on the material morphology, compounding with other materials, and surface modification. It is generally accepted that the ZnO in nanosheet, nanowire, and column structures are more prone to the deformation and piezoelectric potential than its granular counterparts. The degradation performance of hexagonal columnar ZnO is higher than that of flake and spherical ZnO, and hexagonal columnar ZnO shows higher piezoelectric induced bias at the same ultrasonic power.

It has been reported that the electrons are promoted from the VB to the CB of ZnO upon light illumination. Then, the electron-hole pairs are separated by the piezoelectric field, and the separated holes migrate to the surface of ZnO, where the degradation reactions take place (Figs. 4A and B) [33]. The piezoelectric potential of ZnO NRs encourages electrons and holes to interact with dissolved O₂ and OH[−] more favorably, leading to the formation of $\cdot\text{O}_2^-$ and $\cdot\text{OH}$ radicals. The simultaneous renewal of the free charges is also possible by obtaining ultrasonic vibrational energy. A considerable piezoelectric catalysis effect will come from the quick accumulation of reaction products caused by the high concentration of free charges and high strain stress [108]. The catalyst was prepared by growing hexagonal columnar ZnO arrays on a grid-like bottom plate. Different stirring rates were set to observe the degradation of RhB by the catalyst under the conditions of stirring cavitation (Figs. 4C and D) [103]. RhB deteriorates more quickly as the stirring speed is increased.

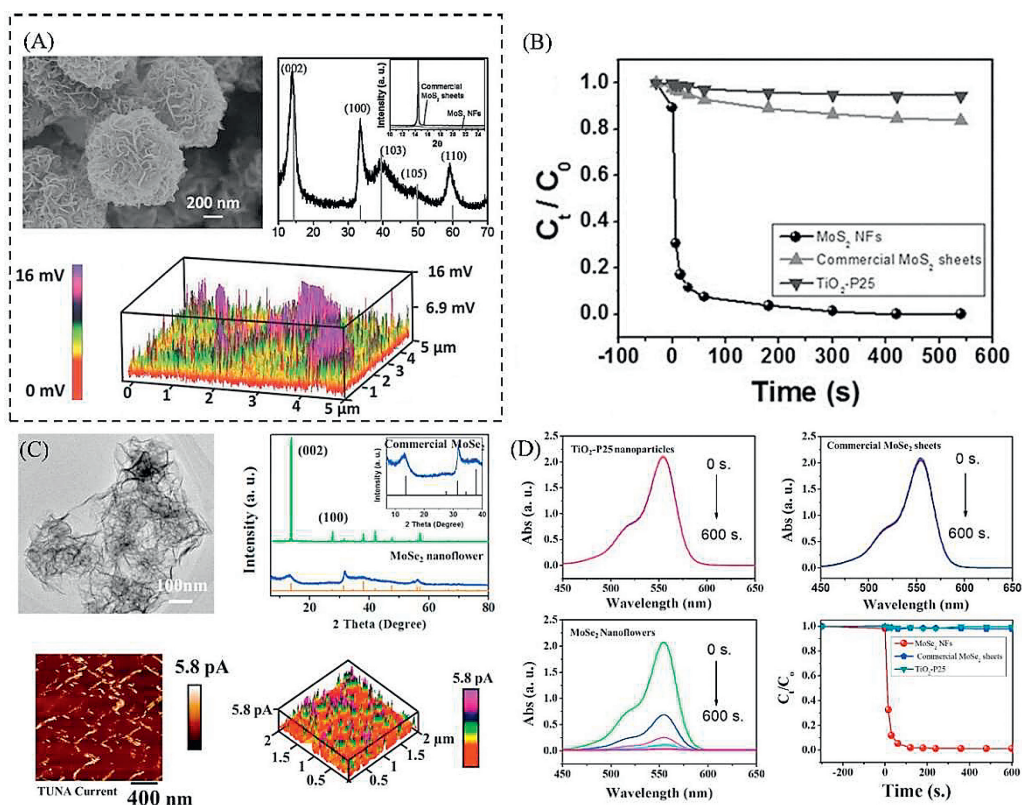


Fig. 3. (A) A huge proportion of nanometals, MoS₂'s XRD, and a piezoelectric potential responsive picture make up the MoS₂ NFs. (B) Utilizing the MoS₂ NFs ultrasonic wave in the dark to measure the degradation ratio of the RhB dye. Copied with permission [54]. Copyright 2016, Wiley Online Library. (C) In the TEM picture of the MoSe₂ nanoflowers, there are many single and few layers, as well as the corresponding TUNA current of the MoSe₂ nanoflowers and a three-dimensional representation of the TUNA current distribution in the MoSe₂ nanoflowers. (D) The development of degrading activity for TiO₂-P25 nanoparticles, commercial MoSe₂ sheets, and synthetic MoSe₂ nanoflowers under ultrasonic-wave vibration in the dark. The evaluation of each catalyst's degradation ratio. Copied with permission [60]. Copyright 2017, Elsevier.

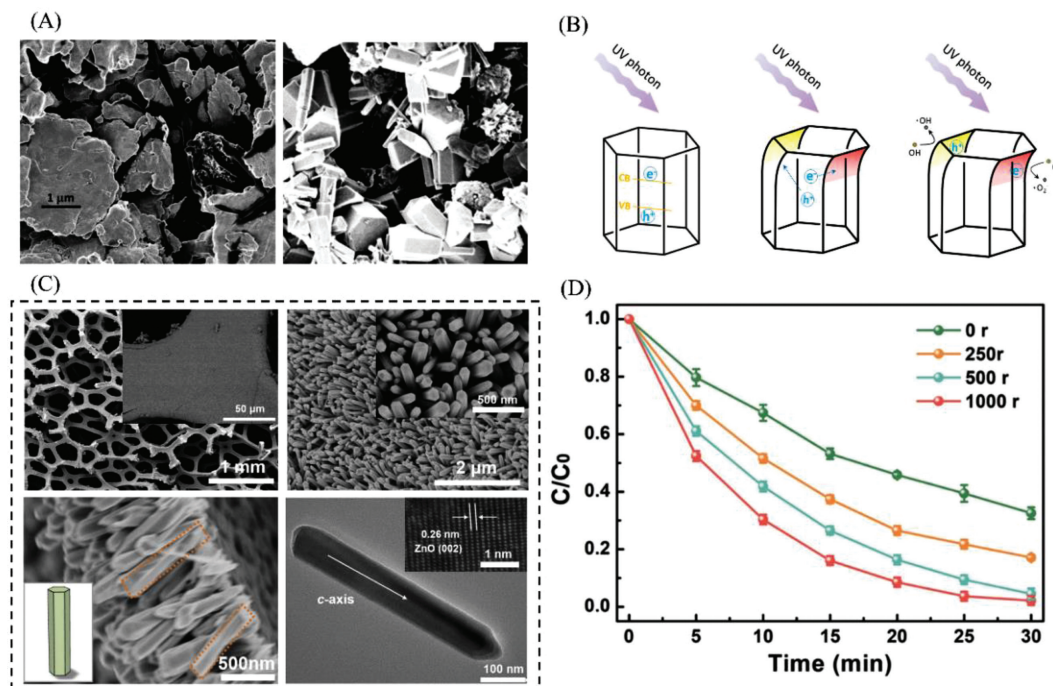


Fig. 4. (A) SEM images of ZnO microstructures: lamellar and hexagonal columnar ZnO. (B) Schematic diagram to illustrate that the electron is promoted from the VB to the CB of ZnO with the presence of light illumination. The presence of the piezoelectric force separates the electron-hole pairs, and the separated charge carriers move to the ZnO surfaces where the degradation events happen. Copied with permission [33]. Copyright 2020, Multidisciplinary Digital Publishing Institute. (C) SEM images of ZnO nanorod array on porous substrate, side view of ZnO NRs, TEM, and HRTEM images of ZnO NRs. (D) Degradation of RhB by the ZnO nanorod array on porous substrate at different stirring speeds. Copied with permission [103]. Copyright 2022, Elsevier.

4. Organic piezoelectric materials for pollutants removal

Lead piezoelectric materials, explanation: lead zirconate titanate (PZT) has become the mainstream of lead-based piezoelectric materials because of its ideal elasticity, dielectric, piezoelectric and other characteristics, in addition to lead metaniobate, lead titanate, *etc.* However, the lead element in these lead-based materials adversely affects humans and ecology. Based on this, researchers have developed lead-free piezoelectric materials: organic and inorganic [109]. Lead-free organic piezoelectric compounds, sometimes known as piezoelectric polymers [109,110]. Included are mostly silicone, epoxy, and other organic piezoelectric compounds [111–113]. Particularly, the advantages of low density, good flexibility, low impedance, and high piezoelectric constant of PVDF and related copolymers and composites have attracted a great deal of research interest in recent years.

4.1. PVDF

The poly(vinylidene fluoride) (PVDF) has gone through a long history since its first developed in 1944, when its piezoelectric properties were discovered, and now it is widely used in our daily lives and scientific research owing to its high piezoelectric coefficient. In PVDF, the electronegativity of F atoms is much higher than that of C and H. Therefore, the charge density is shifted in the crystal structure, resulting in a good piezoelectric effect. PVDF is known to exhibit five phase states, of which α and β are the basic phase states. α phase is not piezoelectric, but β is piezoelectric [114]. The reason is that all the dipole moments in the β phase will point to the same direction and render a higher piezoelectricity [115]. Fascinatingly, previous research divulged that the α phase can be transformed into the β phase by stretching the PVDF film or nanowires [116].

With the use of electrospinning, Zheng *et al.* produced PVDF electrospun fiber immobilized with TCN (PVDF-TCN) that had nanofibers of a consistent size and distribution. The PVDF nanowires' surface had a homogeneous distribution of TCN particles (Fig. 5A) [64]. Under visible light irradiation for 300 min, PVDF electrospun fibers exhibited negligible photocatalytic activity against tetracycline, whereas PVDF-TCN-0.1 g, PVDF-TCN-0.15 g, and PVDF-TCN-0.2 g had much increased photocatalytic activity, as shown in Fig. 5B. In this case, PVDF plays as a carrier in this catalytic system, efficiently dispersing the particulate $\text{TiO}_2\text{@g-C}_3\text{N}_4$ catalyst, increasing the surface area as well as the active sites exposure. The electrons in the TiO_2 CB then trap O_2 on the catalyst surface, generating the active oxygen species $\cdot\text{O}_2^-$, with a small fraction of h^+ generating the strongly oxidizing $\cdot\text{OH}$ with the surrounding H_2O or OH^- (Fig. 5C) [64]. For the oxidation of pollutants, there are also mixtures of traditional piezoelectric materials and photocatalytic materials. To take advantage of PVDF's effect and improve the low point charge and hole separation efficiency of TiO_2 , PVDF and TiO_2 were layered. The PVDF fiber surface is covered by TiO_2 particles having an average particle size of 20 nm in the PVDF- TiO_2 composite, yet the fibers' structural integrity is preserved (Fig. 5D). The experimental findings demonstrated that the piezoelectric catalysis of methylene blue (MB) aqueous solution by PVDF- TiO_2 composite increased the TOC content. The performance test proves that PVDF- TiO_2 not only assist the decolorization of MB, but also render high mineralization rate (Fig. 5E). This work suggests that the piezoelectric effect induces the separation of the free charge generated by UV light from the holes and that reactive oxygen species are generated, thereby improving the efficiency of dye degradation (Fig. 5F).

4.2. g- C_3N_4

Other than PVDF, g- C_3N_4 also has been widely used in environment applications as a piezoelectric material, owes its excellent properties to its 2D lamellar structure [67]. From a crystallographic point of view, piezoelectric crystals must have groups of points with asymmetric centers. Some layered materials are intercalated, peeled into two-dimensional nanosheets and stimulated by stress, where the center of symmetry is broken, resulting in a piezoelectric potential. The piezoelectric potential forces the charge to separate from the holes and pushes the charge to the surface of the material, thereby participating in the redox reactions. Over the past few decades, g- C_3N_4 is favored by researchers owing to its metal-free properties which consists of only two elements, C and N, environmentally friendly, high chemical stability, ease of preparation, and narrow forbidden band width to facilitate the absorption of visible light. However, all things have two sides. Similar to most of the existing photocatalysts, g- C_3N_4 also suffer from low separation efficiency of electron-hole pairs. Some researchers argue that layered materials without piezoelectric properties are piezoelectric in nature in morphology, as layers are more easily deformed than blocks. The production of gas from the thermal breakdown of NH_4Cl and the purging of CN to generate g- C_3N_4 nanoflakes with a crooked orientation and curled edges, with an average thickness of about 2 nm (Fig. 5G). The linear piezoelectricity of g- C_3N_4 was verified by applying voltages of different intensities to lead to local resonance of the material. Ultrasonic vibration for the breakdown of other CPs and simultaneous irradiation were used to examine the pervasiveness of 2D g- C_3N_4 samples such as 2,5-DCP, 2,6-DCP and 4-CP (Figs. 5H and I) [59].

5. Modification methods

Classical piezoelectric materials have been studied for many years with great progress recently. Despite the fact that there are many uses for removing pollutants from water in the field of water treatment, the realization of this technology for industrial application is still a mirage. Generally, single type of piezoelectric material still exhibits some shortcomings in environmental treatment, such as slow degradation efficiency, limited mineralization, and difficult to recycle. The modifications of piezoelectric materials mainly lie in three aspects: (i) changing the morphological structures of the material; (ii) couple with other materials to form composite; and (iii) surface modification of the material. The state-of-the-art modification approaches are summarized and discussed in this review paper.

5.1. Morphology methods

Nanomaterials can take many different forms, such as nanowires, nanosheets, nanospheres, nano-flowers, nanorods, and more. In general, unusual synthesis techniques can be used to create the aforementioned nanostructures. However, for piezoelectric catalytic performance, the susceptibility to deformation under mechanical stress is the key to the finished material. It is well known that for nanoparticles, nanosheets and nanowires are more prone to deformation. In addition to this, there are also materials with nano-microspheres and nanoflower morphologies that have become popular in recent years. The nanosheet material is a two-dimensional structure, and in piezoelectric catalysis, mechanical stresses force the sheet material to crumple, breaking the center of symmetry to produce the piezoelectric effect. In terms of preparation, they can be synthesized directly or obtained by intercalation and exfoliation of the bulk material. Therefore, altering the morphology is one technique for improving the characteristics of piezoelectric materials. However, it is important to note that when

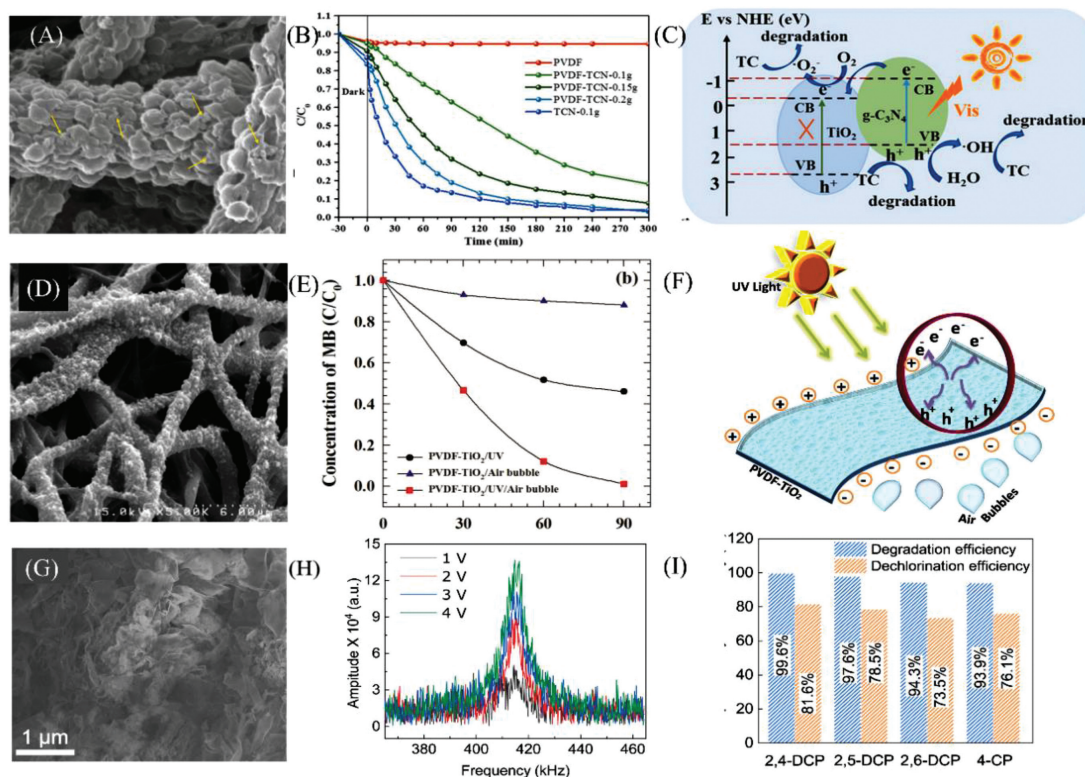


Fig. 5. (A) SEM of PVDF-TCN-0.2 g electrospun fibers. (B) Tetracycline photocatalytic degradation efficiency using TCN, PVDF, and PVDF-TCN. (C) Tetracycline is broken down via a photocatalytic process in PVDF-TCN electrospun fiber. Copied with permission [64]. Copyright 2021, Elsevier. (D) FESEM images of PVDF-TiO₂ hybrid. (E) Degradation of MB using PVDF-TiO₂ hybrid piezo-photocatalysis. (F) Photo-piezoelectric catalytic reaction mechanism. Copied with permission [42]. Copyright 2019, Elsevier. (G) SEM images of 2D g-C₃N₄. (H) Resonant peaks for different applied voltages on 2D g-C₃N₄ nanosheet. (I) Piezocatalytic dechlorination and degradation efficiency of Dichlorophenol (DCP) contaminants by 2D g-C₃N₄. Copied with permission [59]. Copyright 2022, Elsevier.

designing the material form, attention should be paid to the range of stresses that the material can withstand. Too much stress or a material that is too fragile can easily cause irreversible damage to the material, resulting in a reduction in the recyclability of the material.

For nanowire materials, such as ZnO NRs with diameters in the 150–500 nm range, piezoresponse force microscopy (PFM) has shown piezoelectric coefficients ranging from 4.41 pm/V to 7.5 pm/V, while the values vary from 14.3 pm/V to 26.7 pm/V for ZnO nanobelts with tens of nm in thickness. As opposed to the bulk value of 12.4 pm/V, a resonance shift approach has shown a value as high as 12,000 pm/V for a ZnO nanowire with a diameter of 230 nm [67,117,118]. The amount of the piezoelectric coefficient is directly proportional to the rate of polarization per unit volume of the material, and both the charge redistribution in the axial direction of the nanowires and the rearranging of the atoms reduce the polarization overall. By comparing nanowire and bulk materials, the contraction of surface relaxation in the radial direction leads to an enhanced polarization per unit volume and thus inside the crystal of the nanowire morphology [117]. The second is the effect of particle size on the piezoelectric coefficient [119]. Systems with mixed bonding characteristics become more ionized as the grain size decreases. It is hypothesized that ionic bonds are spherically symmetrical, so the loss of covalency in the bonding features will show a tendency to induce a high degree of lattice symmetry. The stronger the lattice's symmetry and the less apparent the piezoelectricity, the smaller the grain size [119,120].

Researchers fabricated Na_{0.5}Bi_{0.5}TiO₃ (NBTO) with different morphologies by template hydrothermal method and hydrothermal method [121,122]. As shown in Fig. 6, there are three morphologies of NBTO, which are NBTO nanowires [1], NBTO nanocrystals and

NBTO nanospheres (SEM images in Figs. 6A, D and G) [1,24,25]. The EDS element mappings in Figs. 6B, E and H show that the elements Na, Bi, Ti, and O are uniformly distributed in NBTO. Based on the degradation tests, NBTO nanowires completely degraded 5 mg/L of RhB within 80 min [1], NBTO nanocrystals degrade 98% of RhB in 180 min by synergistic photo-piezoelectric action, whereas the NBTO nanoparticles achieved a maximum degradation rate of 0.022 min⁻¹ (Figs. 6C, F and I) [1,24,25]. In summary, the morphology design of piezoelectric catalytic materials can be based on the ease of deformation. The general rule is 1D morphology > 2D morphology > 3D morphology.

5.2. Composites with heterojunctions

In piezoelectric catalysis, the piezoelectric potential can drive the separation of electrons from holes [123,124]. However, carrier translation shields the spontaneous polarization field, leading to a loss of carrier separation driving force. A strong piezoelectric polarization field with a high carrier concentration is needed to reduce the mechanical energy consumption and increase optical energy utilization [123]. The building of heterojunctions has recently been a hot study subject in the realm of piezo-photocatalysis because the heterojunction interface can considerably increase the carrier separation efficiency. Heterojunction, the interface region formed when two different semiconductors are in contact with each other. The usual conditions for forming a heterojunction are that the two semiconductors have similar crystal structures, similar atomic spacing and thermal expansion coefficients. For piezoelectric materials, the heterogeneous structure can effectively separate charge and hole, reduce the compound rate of both, and improve the electrical conductivity and electron-hole utilization [125–130].

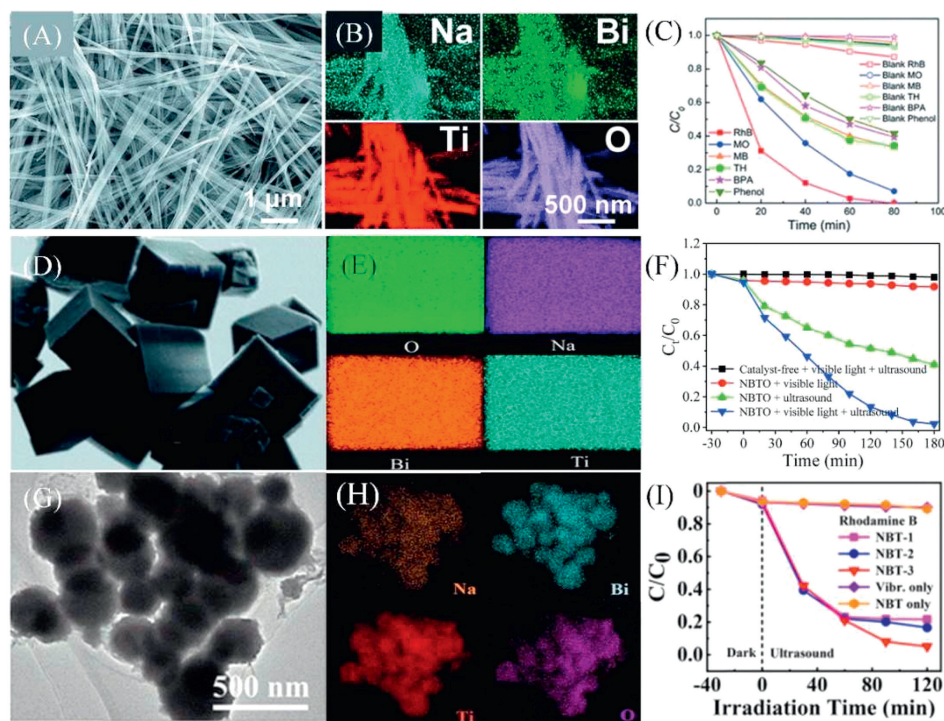


Fig. 6. (A) SEM image, (B) EDS mapping, and (C) piezo-catalytic degradation dynamic curves of $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ nanowires. Copied with permission [1]. Copyright 2021, Royal Society of Chemistry. (D) SEM image, (E) EDS mapping and (F) piezo-catalytic degradation dynamic curves of $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ μm cuboid. Copied with permission [25]. Copyright 2020, Royal Society of Chemistry. (G) TEM image, (H) EDS mapping, and (I) piezo-catalytic degradation curves of $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$. Copied with permission [24]. Copyright 2022, Elsevier.

In the case of piezoelectric materials, the two materials are usually compounded to form a heterojunction structure at the interface contact. There are many works on the preparation of heterojunction composites [131]. Here, we classify the currently common piezoelectric heterojunction materials into two categories: (i) non-piezoelectric substrate materials with piezoelectric materials, (ii) dual piezoelectric materials (include homogeneous heterogeneous junctions p-p, n-n, and non-homogeneous heterogeneous junctions p-n) [132,133].

Carbon is a commonly studied non-piezoelectric material to compound with piezoelectric materials. The carbon source is mostly carbon nanotubes, carbon cloth and other materials such as monolayer, multilayer graphene, biochar material. Composite with carbon materials can effectively improve the electrical conductivity of the material. $\text{Mo}_2\text{S}_3/\text{C}$ heterojunction materials can be prepared by template hydrothermal method. SEM image show that the $\text{Mo}_2\text{S}_3/\text{C}$ is in the form of a large number of overlapping ultrathin nanosheets with crystal lattice spacing of about 0.53~0.85 nm, and more cracks and folds can be seen on the surface of the material (Figs. 7A and B). Performance tests showed that the degradation rate of contaminants was effectively enhanced by ultrasonic irradiation of $\text{Mo}_2\text{S}_3/\text{C}$ in the Fenton system (Fig. 7C) [56].

The second category refers to the composite consists of two different types of piezoelectric materials. The representative composite is $\text{BaTiO}_3/\text{La}_2\text{Ti}_2\text{O}_7$ (BTO/LTO) heterostructure. The surface potential of the composite, which is much higher than that of a single BTO or LTO, indicates that the presence of a polarization electric field can effectively regulate the carrier migration and separation at the heterojunction interface. The n-n heterojunction is formed between BTO and LTO [22]. The BTO and LTO composite can be formed by a facile microwave hydrothermal method. LTO nanosheets are grown *in situ* on BTO nanorods to form a rough surface (Figs. 7D and E). Performance testing under the same ul-

trasonic conditions revealed that the composite degraded contaminant CIP more efficiently than the single incomposite BTO and LTO (Fig. 7F) [22]. In addition to this, there are also multiple composite piezoelectric materials. The p-n type heterojunction photocatalytic material $\text{Ti}_{23}\text{-oxo-cluster}/\text{BaTiO}_3/\text{CuS}$ also exhibited excellent performance in pollutants degradation under the conditions of photopiezoelectric synergy. As demonstrated from SEM and TEM photos, the CuS nanospheres with smooth flower-like morphology became rather rough after the composite deposition of BaTiO_3 material (Figs. 7G and H). From the degradation tests, the ternary composite achieved ~100% degradation of the pollutants, which signifies almost complete degradation (Fig. 7I) [134]. Besides depositing CoO_x nanoparticles on the surface of BFO nanodiscs, the formed heterojunctions can achieve the same effect of promoting electron-hole separation. It is noteworthy that CoO_x nanoparticles are often used as accelerators in catalysis and also as electrons trapping sites [135].

5.3. Surface modification

A low piezo-catalytic reactivity is caused by the majority of piezoelectric materials' poor electric conducting qualities and restricted concentration of free carriers, which severely limit the movement of charge carriers. Surface modification is to artificially create various defects as these defects can effectively trap the charge carriers. There are two main methods of surface modification, which are loading the surface of the material with precious metals and introducing defective spots on the surface of the material [12].

Noble metals (Ag, Au, and Pt) can be added to the surface of piezoelectric catalysts to increase their catalytic activity because the Schottky barriers that develop at their interface act as electron traps and hasten the separation of electron-hole pairs. On the other

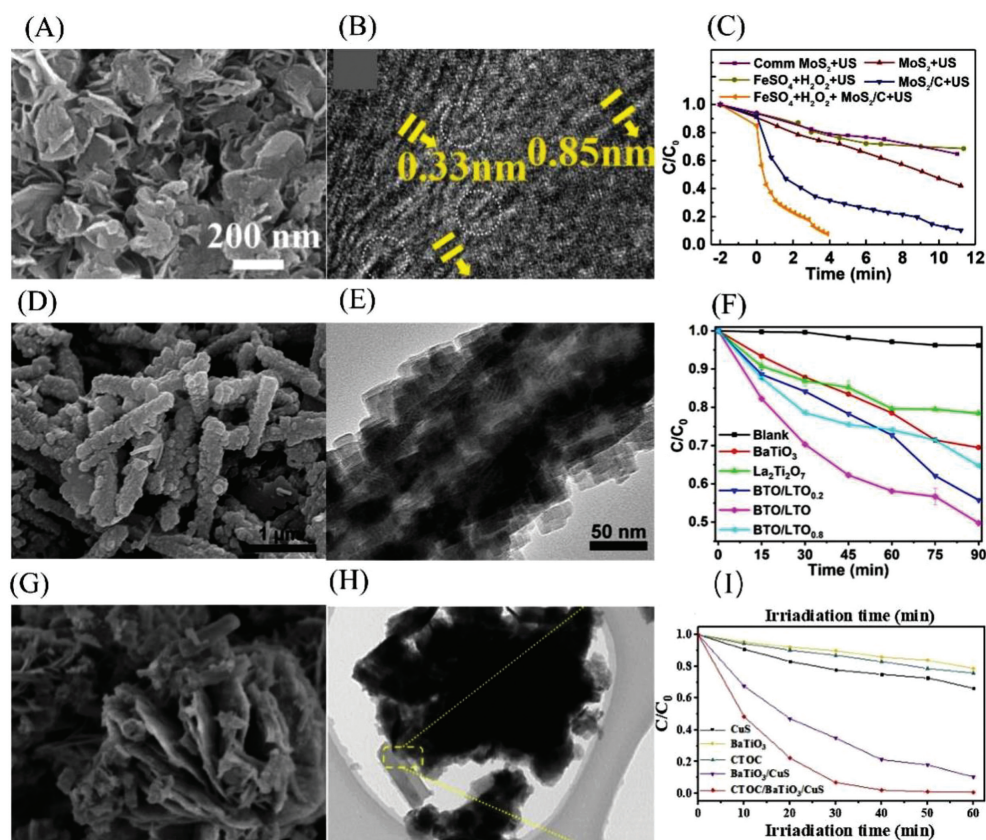


Fig. 7. (A) SEM image of MoS₂/C and (B) TEM image of MoS₂/C. (C) Piezocatalytic degradation kinetics of imidacloprid (IMD) with different catalysts under ultrasonic irradiation. Copied with permission [56]. Copyright 2020, Elsevier. (D) SEM image and (E) TEM image of BTO/LTO composites. (F) Piezo-photocatalysis with ultrasound conditions of 210W and 40 kHz. Copied with permission [22]. Copyright 2021, Elsevier. (G) SEM image and (H) TEM image of CTOC/BaTiO₃/CuS. (I) Degradation activity of photo-piezoelectric catalysis. Copied with permission [134]. Copyright 2021, Elsevier.

hand, Bi nanoparticles are also reported to deposit on a smooth and flat BiOCl nanosheet. The Bi/BiOCl composite piezoelectric material with irregular morphology can be obtained. For deposition, the Bi nanoparticles are primarily focused on the nanosheet edges (Figs. 8A and B). The degradation of carbamazepine showed an increasing and then decreasing trend with the increase of Bi deposition (Fig. 8C) [40]. Researchers have discovered that the dynamic piezoelectric field of the piezoelectric material increases in intensity and frequency as oxygen vacancies (OVs) content increases. OVs are the vacancies formed by the detachment of oxygen atoms (oxygen ions) from the lattice in metal oxides or other oxygen-containing compounds, resulting in the lack of oxygen. ZnO microspheres with various OV concentrations are generated under various atmospheric conditions. Crossed nanosheets and irregular polygonal holes generated on the exterior of ZnO microspheres are visible in the SEM image of the V-ZnO microspheres (Fig. 8D) [27]. The XRD of ZnO with different OVs contents did not show any significant difference. The photocatalytic degradation efficiencies of MO, RhB and AO7 by A-ZnO, H-ZnO, and V-ZnO microsphere (Fig. 8E). The ZnO microspheres' ability to photo-catalyze better and exhibit the piezoelectric catalytic property depends on the piezoelectric field and higher OVs concentration. ZnO porous microspheres may destroy organic contaminants under vacuum conditions, which considerably encourages the use of ZnO porous catalysts (Fig. 8F) [27].

In addition to introducing defects on the surface of the material, single-atom catalysts have become increasingly popular. A non-uniform aggregate of hundreds or thousands of metal atoms may result from the modification of materials by depositing, and only a tiny portion of these aggregates will be exposed to the re-

actants due to the low metal atom usage. So, increasing the metal atom utilization of a catalyst is particularly important [136]. Fe1-MoS₂ material was synthesized via a dipping calcination method. HAADF-STEM, which demonstrates the effective deposition of iron single atoms on the surface of MoS₂, can be used to visualize the existence of stronger white photoelectrons on the material's surface (Fig. 8G). Since there is no difference in the XRD patterns before and after deposition, this indicates that the iron is not compounded with MoS₂ in the form of crystals (Fig. 8H). Degradation experiments showed that the contaminant Metronidazole (MTZ) could be effectively degraded with the addition of catalyst and PMS (Fig. 8I). This is resulted from the isolated Fe atomic sites to play several roles: (i) improved the piezoelectric polarization of MoS₂; (ii) accelerated piezoelectric charge separation of MoS₂; and (iii) enhanced PMS activation for water purification in the piezo-catalytic process [137].

6. Summary and perspectives

In summary, this article summarizes the research progress in the last two years concerning piezoelectric materials for wastewater treatment under ultrasonic cavitation conditions. Firstly, the piezoelectric principle of piezoelectric materials, the intrinsic structure of classical materials, and the reasons affecting the performance of piezoelectric materials are introduced. Subsequently, the commonly used piezoelectric driving method and ultrasonic cavitation are discussed and the cavitation principle is briefly described. Following that, some classical piezoelectric materials for pollutants degradation are classified and discussed. On the basis of these materials, some common modification techniques such

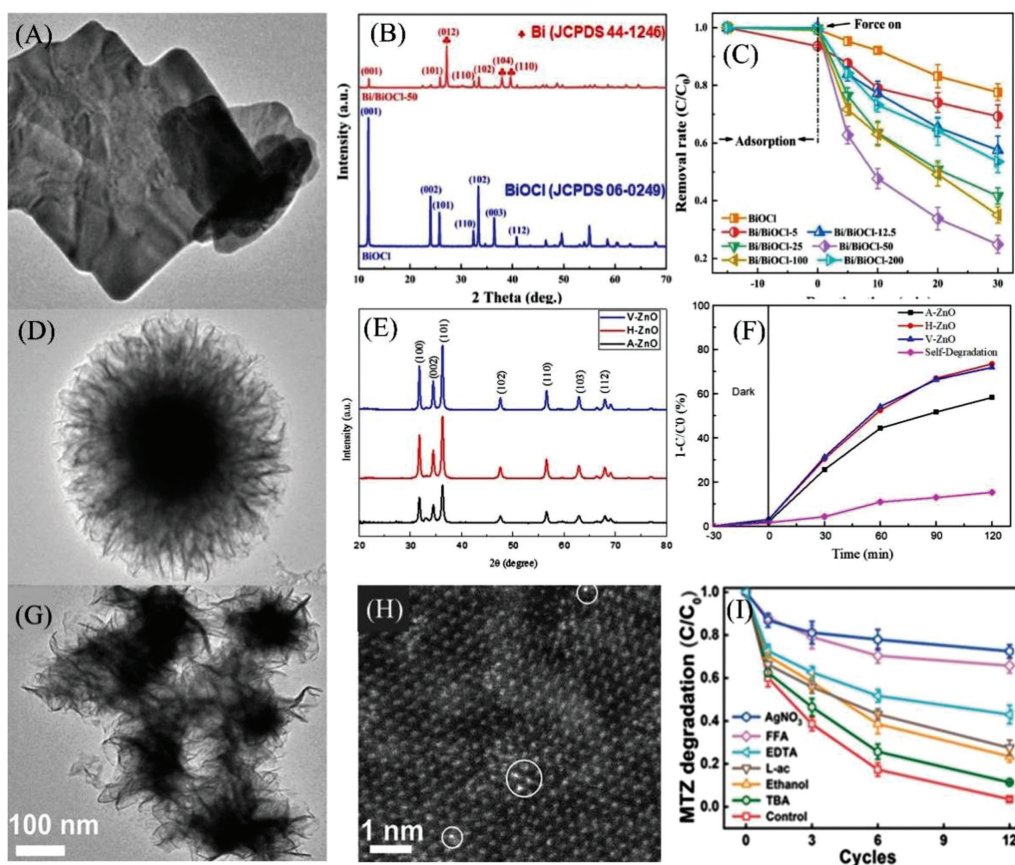


Fig. 8. (A) TEM images of Bi/BiOCl nanocomposites. (B) XRD patterns of the prepared samples. (C) Piezo-catalytic degradation efficiencies. Copied with permission [40]. Copyright 2021, Elsevier. (D) TEM images of V-ZnO nanosheet microsphere. (E) XRD patterns of ZnO nanosheet microsphere. (F) Under ultraviolet radiation, A-ZnO, H-ZnO, and V-ZnO microspheres degrade dye. Copied with permission [27]. Copyright 2020, Springer Link. (G) TEM and (H) HAADF-STEM images of Fe₁-MoS₂. (I) A comparison of the effectiveness of MTZ deterioration in various systems. Copied with permission [137]. Copyright 2022, Wiley Online Library.

as heterostructure composites and surface doping are briefly described. Finally, the practical applications of piezoelectric materials in the treatment of water environments are described, such as degradation of dye wastewater, antibiotic wastewater [138].

Piezoelectric materials collect the changing stress in the environment and convert it into chemical energy to participate in the redox reactions of organic pollutants in water bodies, thus achieving pollutants degradation and thereby purifying water bodies. However, in practical applications, there are still several problems such as insufficient driving force of piezoelectric catalysis, incomplete catalytic degradation effect, and still pollutant intermediates that have to be addressed. Meanwhile, the limited carrier utilization efficiency in the catalytic process also greatly hampers the degradation efficiency. In view of these, piezoelectric catalysis still has great potential for development.

Future advancements in piezoelectric catalysis can concentrate on three different areas. The initial consideration was the catalyst for piezoelectric catalysis. In practical applications, the driving force of natural water body comes from the oscillation of water flow, and in the laboratory, the cavitation effect from ultrasonic cavitation is generally employed. Choosing a suitable driving force can better induce the piezoelectric effect of the material and improve the efficiency of converting mechanical energy into chemical energy. The second aspect focuses on the materials and material modification. More rational material structures should be designed to improve the utilization of material carriers. Last but not least, the third aspect involves characterization tools and theoretical calculations. Since piezoelectric catalysis is still an emerging research

field, the utilization of appropriate characterization tools and theoretical calculations are beneficial to deepen our understanding for the design and applications of piezoelectric materials.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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