

Review

Ultrasonic-Assisted Piezoelectricity by Barium Titanate Materials: Multi-Domain Application and Mechanism Exploration

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Abstract: Barium titanate-based materials serve as highly efficient catalysts in the realm of piezoelectric ceramics. With the aid of ultrasound, these piezoelectric ceramics have been widely used and exhibit remarkable potential in advancing wireless, precise control of therapeutic technologies, medical applications, and environmental pollutant control. There is a growing interest in leveraging these materials to develop innovative methods for degrading emerging contaminants, but limited articles have explored the feasibility of this application. As such, we review the progress of research in ultrasonic-assisted piezoelectric catalysis by barium titanate materials (BTO&US) and discuss the underlying catalytic mechanisms in various application scenarios. In addition, we outline its future research directions, focusing on crystal structure optimization, electronic density regulation, medium, and catalytic applications for the degradation of emerging contaminants. This review on BTO&US offers novel ideas and methodologies that contribute to the development and application of piezoelectric ceramics.

Keywords: BaTiO₃; emerging contaminants degradation; piezoelectric catalysis; radical; ultrasound

1. Introduction

Piezoelectric ceramic, as an essential energy-conversion material, enables the transformation of mechanical energy into electricity [1–5]. With the assistance of mechanic vibration, the piezoelectric potential is generated within the piezoelectric ceramic. Subsequently, electrons are excited from the valence band to the conduction band, initiating redox reactions on the material's surface [6]. This process, referred to as piezoelectric catalysis, represents a relatively recent technological advancement in recent years [7,8]. Barium titanate (BaTiO₃, BTO) is a pioneer of piezoelectric ceramics, possessing a significant piezoelectric catalytic capacity [9–11] attributed to its polarization at atmospheric pressure. Furthermore, being lead-free and environmentally friendly, it is highly suitable for modern applications compared to similar lead-containing materials. The piezoelectric properties of barium titanate can be stimulated by many external stimuli, including extrusion, ball milling, impact, and ultrasonic vibration (US) [12]. Compared to other vibration modes, ultrasonic vibration stands out due to its wireless excitation and deep energy penetration characteristics, making it highly compatible with collaborative applications with piezoelectric materials. Hence, ultrasonic-assisted piezoelectricity by barium titanate materials (BTO&US) represents a wireless excitation method to enhance the piezoelectric catalytic capacity of BTO. This ingenious approach effectively combines the strengths of both techniques, offering remarkable advantages, including wireless control, universality, precise control, and biosafety. At present, BTO&US is used in medicine, biology, chemistry, etc. It has been researched in more than 10 aspects (Figure 1). In addition, researchers have leveraged this method to develop innovative cancer treatment protocols, wireless medical materials, and novel approaches



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for pollutant treatment. The potential of this method extends far beyond these applications and offers opportunities for exploration in new fields and addressing emerging challenges.

Emerging contaminants like algal toxins and antibiotics pose a growing threat to humans and aquatic life. Traditional physical and chemical methods fail to convert them into beneficial compounds [13], and biological methods are relatively ineffective in addressing trace organic pollutants [14]. Free radicals have been proven to be applicable in this field. Given the diverse array of free radicals generated during the piezoelectric catalysis process [15,16], there is anticipation for BTO&US to exhibit performance in this domain. It is evident that the study of BTO&US, encompassing mechanisms in application, practical research domains, and methods for material optimization, holds significant value in broadening treatment methodologies for emerging contaminants. However, a systematic summary and analysis of the research field and application mechanism of BTO&US have not been undertaken to date. In terms of mechanisms, researchers have different depths and angles of mechanism exploration in different works, but significant limitations persist. In terms of the research field, although some researchers have made attempts to degrade emerging contaminants using this method, the scope of research remains limited, leaving considerable untapped potential for further exploration and application.

This paper summarizes the research progress of BTO&US, presenting its current status in various fields and demonstrating its feasibility in degrading emerging contaminants. Additionally, it provides a summary of the catalytic and application mechanisms of BTO&US. The catalytic mechanism is explained in general through three stages: ‘absorption of vibration’, ‘production of active substances’, and ‘application in different scenarios’. Characterization analysis methods and potential research directions for further exploration are provided for each stage. Lastly, the paper discusses the prospects of this method, offering insights into new trends in the development of BTO&US, which will aid researchers in gaining a deeper and more intuitive understanding of the mechanism and potential of this approach.

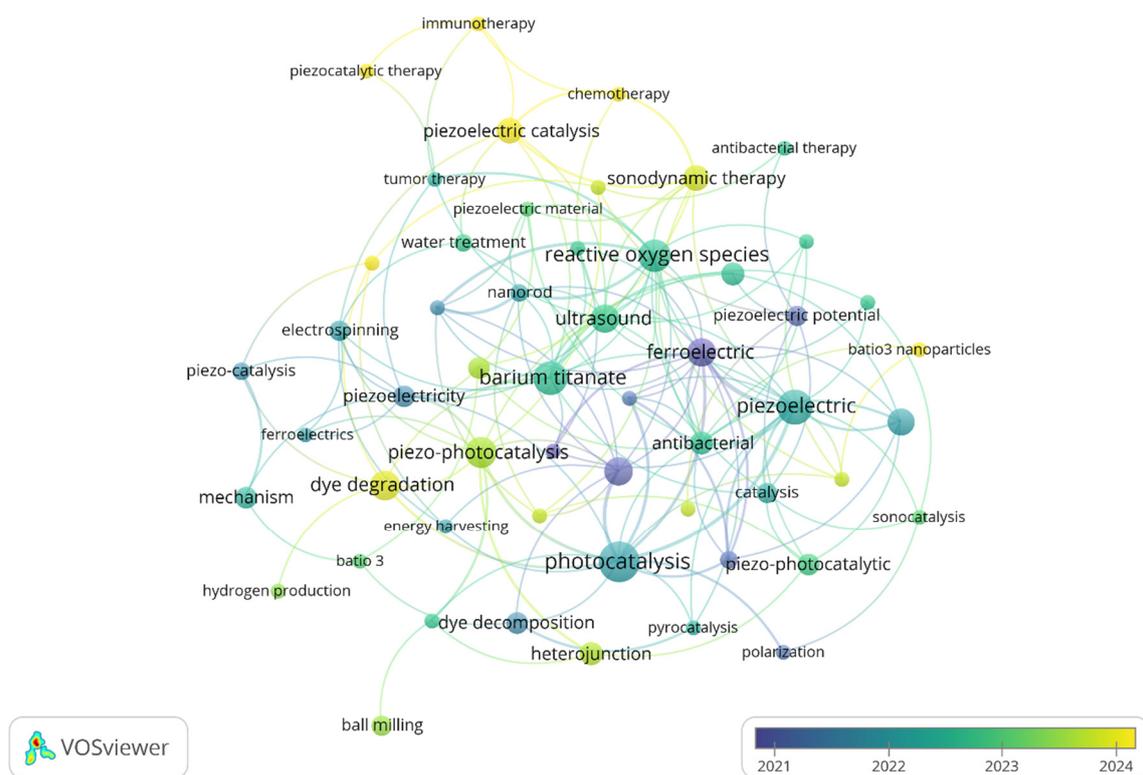


Figure 1. Publications featured with the theme of BTO&US retrieved from the Web of Science (WoS).

2. Current Research Status of BTO&US in the Main Fields

2.1. Medical Application

2.1.1. Cancer Treatment

Cancer treatment remains a challenging problem, with patients hoping for effective and non-invasive treatment alternatives to alleviate the pain associated with radiation therapy, invasive surgeries, and other

procedures. The power and frequency of ultrasound in BTO&US are adjustable, enabling the expansion of the wireless treatment scheme to areas within the organism that are difficult to access. Different kinds of tumor cells can be tackled by it. In pure barium titanate, the tetragonal BTO possesses the characteristic of generating polarization in the internal electric field after exposure to ultrasound. Leveraging this property, a local tumor eradication method with high biosafety has been devised. This method generates $\cdot\text{O}_2^-$ or $\cdot\text{OH}$ to eliminate 4T1 breast cancer cells and eradicate the tumor effectively [17]. Tetragonal BTO nanoparticles are optimal for localized tumor therapy via ultrasound-triggered ROS, while ultrasmall PEGylated BTO nanoparticles are superior for systemic treatment of metastatic triple-negative breast cancer through hypoxia alleviation (O_2 generation) and metastasis suppression. What is more, ultra-small BTO nanoparticles also offer a new method for treating triple-negative breast cancer. The piezoelectric effect decomposes water, generating reactive oxygen species (ROS) and oxygen. This process alleviates the expression of hypoxia-inducible factor-1 α in the solid tumor microenvironment, ultimately inducing apoptosis of tumor cells [18]. Moreover, Zhan et al. experimentally demonstrated that BTO can produce internal radio stimulation, resulting in a notable inhibition of tumor cell proliferation [19]. Therefore, the research group proposed a non-invasive treatment approach for triple-negative breast cancer, highlighting its advantage of causing minimal side effects. In addition, the composite materials with BTO as the core can realize complementary advantages and lead to significant performance enhancements. An example is the utilization of piezomagnetic nanoparticles, achieved by combining BTO with superparamagnetic iron oxide nanoparticles. These composites showcase exceptional ferroelectric and conductive properties, enabling the eradication of residual osteosarcoma cells. They hold great potential for comprehensive application in postoperative treatment for osteosarcoma [20]. Besides, the antibody-functionalized BTO nanoparticles have dual targeting ability. BTO functionalized with transferrin receptor antibodies effectively inhibits the proliferation of glioblastoma multiforme and promotes their apoptosis [21]. BTO functionalized with anti-HER2 antibody exhibits the ability to inhibit the proliferation of breast cancer cells through chronic piezoelectric stimulation while leaving the activity of healthy cells unaffected [22]. Likewise, combining BTO with various metals in different configurations enhances its catalytic capability. Take the bismuth-doped anoxic BTO, for example; it can effectively inhibit the growth of ovarian cancer. Another example is the excellent performance of $\text{Cu}_2\text{-xO-BaTiO}_3$ piezoelectric heterostructure in treating murine refractory breast cancer [23,24].

2.1.2. Auxiliary Medical Materials

BTO material possesses a level of biological safety, ensuring that it does not induce inflammation or thermal damage effects after being activated by ultrasound [17,25]. This is the reason it can serve as a material for auxiliary medicine. Materials with BTO as the central component have found extensive application in the field of biomedicine [26]. Moreover, several research cases highlight its application in piezoelectric catalytic therapy. The adjustable properties of ultrasound further enhance the excellence of barium titanate in this field. Utilizing ultrasound-mediated barium titanate nanoparticles (BTO NPs) can create localized electromagnetic fields. This capability allows for the modulation of the inferior right ganglionated plexus of the heart and a reduction in ventricular rate during rapid atrial pacing induced atrial fibrillation. This makes barium titanate a promising material for auxiliary nerve regulation and offers valuable insights for investigating treatment approaches to control atrial fibrillation rates [25]. In addition, the presence of BTO NPs within the polymer film allows for controlled drug release through ultrasound adjustments. The piezoelectric effect of BTO directly influences the polyelectrolyte structure of the polymer, enabling this clever integration. This ingenious combination opens up a new way to refine intelligent, responsive materials and offers a novel approach to modulate the piezoelectric capacity of BTO through ultrasound excitation precisely [27]. Finally, the wireless control capabilities of BTO&US enable it to play a role in non-invasive therapy. Leveraging this advantage, a PVDF/BTO composite film is employed for cell harvesting, effectively regulating cell adhesion and separating cells from adhesion carriers in a wireless manner. Remarkably, this approach also sustains cell vitality and proliferation. This study broadens the application of BTO&US in regenerative medicine, highlighting the potential of barium titanate-based materials to evolve into a non-invasive, precise, and controllable platform [28].

2.2. Application in Biology

2.2.1. Controlled Cellular Responses to Piezoelectric Stimulation

BTO&US encompasses many subdivisions within the domain of cell control. Diverse schemes have emerged based on varying cellular responses to piezoelectric stimulation. Table 1 presents a snapshot of the research progress in this area. The composite materials predominantly composed of barium titanate exhibit enhanced

efficacy. In addition, this method enables wireless control of biological processes within cells, offering selectivity, controllability, and broad applicability as distinct advantages.

Table 1. Application of BTO&US in the field of cell control.

No.	Term	Content	References
1	Cell stimulation	1. The tetragonal BTO nanoparticles, when exposed to ultrasound, have the capability to induce a Ca ²⁺ influx. This influx can stimulate neurons, promoting cell growth and differentiation.	[29]
2		2. The colloidal TiO ₂ -BTO composite nanorods, under the influence of wireless ultrasound, can generate ROS within the cell, inducing cell stress or even cell death.	[30]
3		3. Piezoelectric BTO nanoparticles can stimulate neuron cells, with the aid of ultrasound, selectively modulating the activity of neuronal subpopulations in the central nervous system of animals.	[31]
4	Cell differentiation	1. The three-dimensional piezoelectric structure fabricated by BTO combined with Two-Photon Lithography technology, under ultrasound assistance, effectively promotes the differentiation of SaOS-2 osteoblasts, demonstrating an approximately 5-fold enhancement compared to the control group.	[32]
5		2. The bio-ink formulated with BTO nanoparticles, in the presence of ultrasound, facilitates the differentiation of muscle cells.	[33]
6		3. The polymer/BTO composite, aided by ultrasound, can stimulate the growth and differentiation of SH-SY5Y neuroblastoma cells.	[34]
7	Cell proliferation	1. BTO as a coating with Ti ₆ Al ₄ V scaffold, with the assistance of ultrasound, can activate more mitochondria and promote cell proliferation at the initial stage.	[35]
8		2. BTO flexible piezoelectric membrane prepared via electrospinning, with the aid of ultrasound, can positively impact wound healing. The wound closure rate in the animal model was enhanced from 64.15% in the control group to 100% on day 9 with the ultrasound-activated piezoelectric membrane.	[36]

2.2.2. Antimicrobial Application

Research and application of antibacterial materials currently represent a prominent research focus. BTO&US has also made significant advancements in this field. Feng et al. pioneered the piezoelectric effect for disinfection, showcasing the bactericidal potential of BTO&US [37]. Tetragonal BaTiO₃ with nanometer or micrometer size exhibited the ability to effectively eliminate *Escherichia coli* with ultrasound assistance, meeting the required inactivation thresholds for industrial antimicrobial agents (2 logs). Their study revealed that the sterilization process involves a combination of physical damage and chemical oxidation. This shows the synergistic effects of piezoelectric catalysis and ultrasonic mechanical destruction on cell inactivation when employing this sterilization method. In addition, research in this domain has focused notably on composite materials. Rapid in situ sterilization and bone repair stand out as crucial approaches to address infections in bone defects. Within the BTO&US system, sulfur-doped BTO emerges as a promising solution meeting both these requirements. It exhibits outstanding antibacterial properties and proves highly effective in eradicating *Staphylococcus aureus*. Simultaneously, electrical signals generated during the piezoelectric process stimulate the osteogenesis induction of human bone marrow mesenchymal stem cells. This method successfully cured the rat tibia bone defects with depressed inflammation infected by these bacteria [38]. Au@BTO also shows high antibacterial efficiency against representative Gram-negative bacteria (*Escherichia coli*) and Gram-positive bacteria (*Staphylococcus aureus*) [39]. The TiO₂/BTO/Au multilayer coaxial heterostructure nanorod array serves as an effective antibacterial coating, achieving an impressive removal rate of 99.9% for both gram-negative bacteria *Escherichia coli* and gram-positive bacteria *Staphylococcus aureus* under simulated sunlight. In addition, it can accelerate skin tissue regeneration and promote wound healing [40]. Photodynamic light-activated coatings (TiO₂/BTO/Au) target superficial infections via ROS under UV/visible light, while piezoelectric sonodynamic ultrasound-activated nanocomposites (Au@BTO) enable deep-tissue antibacterial therapy and regeneration via ultrasound-triggered redox modulation. Besides, Ag-BTO can be used to inactivate *Escherichia coli*. Real water samples can be used as the treatment object to inactivate cultural bacteria within 5 min and kill alive but unculturable bacteria within 20 min. These results substantiate the feasibility of this method in dealing with real water samples [41].

2.3. Application in Environmental Remediation

2.3.1. Degradation of Model Organic Compounds

The free radicals generated during the piezoelectric catalytic process of BTO&US are potent tools for degrading organic substances and possess broad applicability. Numerous studies have shown that this can eventually mineralize macromolecular organic matter into smaller molecules, such as CO₂ and H₂O [42]. It represents an advanced oxidation technology that is highly environmentally friendly. It can not only be applied in water purification and pollution control but also daily life, such as the application scenario of piezoelectric catalytic teeth whitening [43]. Dyes serve as suitable options for researchers to validate material properties. Methyl orange (MO) and Rhodamine B (RhB) are commonly used dyes in this field. In the BTO&US studies, more than 20 different organics have been degraded. Table 2 presents the associated degradation efficiencies. In the degradation of MO, the capacity of BTO varies with different morphologies and sizes. BTO nanowires and nanoparticles exhibit activity, but materials with smaller diameters or sizes display superior performance [44,45]. Modifying silver nanoparticles on BTO or surface modification with silver nanoparticles can further enhance this ability [9,46]. Additionally, the BTO/CDs composite can enhance the piezoelectric photocatalysis ability through a synergetic effect [47]. The hybrid nanofibers constructed by BTO and carbon dioxide can also considerably upgrade the photocatalytic capability of piezoelectric materials. Notably, this method's degradation efficiency is seven times greater than that of traditional piezoelectric materials [48]. As shown in Figure 2a, a performance comparison of MO degradation is presented. Regarding the degradation of RhB, configurations like the tetragonal phase ladder, silver-loaded structures, polymer composites, hierarchical core-shell nanostructures, and multiphase BTO all demonstrate substantial degradation efficiency [49–52]. Tetragonal terrace-like dendrites (91% degradation, $k = 13 \times 10^{-3} \text{ min}^{-1}$), Ag-BaTiO₃ (83% degradation via piezo-photocatalytic synergy), and carbon-shelled BTO@C core-shell (100% degradation, $k = 35.85 \times 10^{-3} \text{ min}^{-1}$) exhibit optimal RhB removal under ultrasound/light, excelling in stability, plasmonic enhancement, and adsorption-oxygen vacancy synergy respectively. This interaction between materials causes changes in oxygen vacancy concentration and ferroelectric response. Of course, some composites have obtained many unique properties. For instance, the electrospun nanofiber structure of BTO possesses a large specific surface area, fine grain size, and variability [53]. Another example is that the BTO polydimethylsiloxane composite porous foam catalyst can significantly reduce secondary pollution and exhibit excellent repeatability [15]. As shown in Figure 2b, the performance comparison of RhB degradation is demonstrated. In the degradation of other common organic pollutants, BTO-based composites exhibit remarkable efficacy. For instance, BTO nanosheets with a high exposure {001} surface can degrade MO, Methylene Blue (MB), RhB, Phenol, Tetracycline Hydrochloride (TH), and Bisphenol A (BPA) [12]. BTO/ZnO composite can degrade MB, MO, Rh B, etc.; levofloxacin antibiotics (LEVO) can also be effectively degraded [54]. BTO NPs can degrade Congo Red, TH, RhB, MB, MO, etc., through a piezoelectric-Fenton synergistic effect. Li-doped BTO can degrade anionic dyes like MB and malachite green [7]. The flexible BTO nanofiber membrane efficiently degrades MB without complex recovery operations, demonstrating excellent repeatability [55]. When BTO&US is supplemented with an additional light source, it demonstrates efficient degradation of trimethoxy pyrimidine (TMP) and nitropyram (NTP) [10,56].

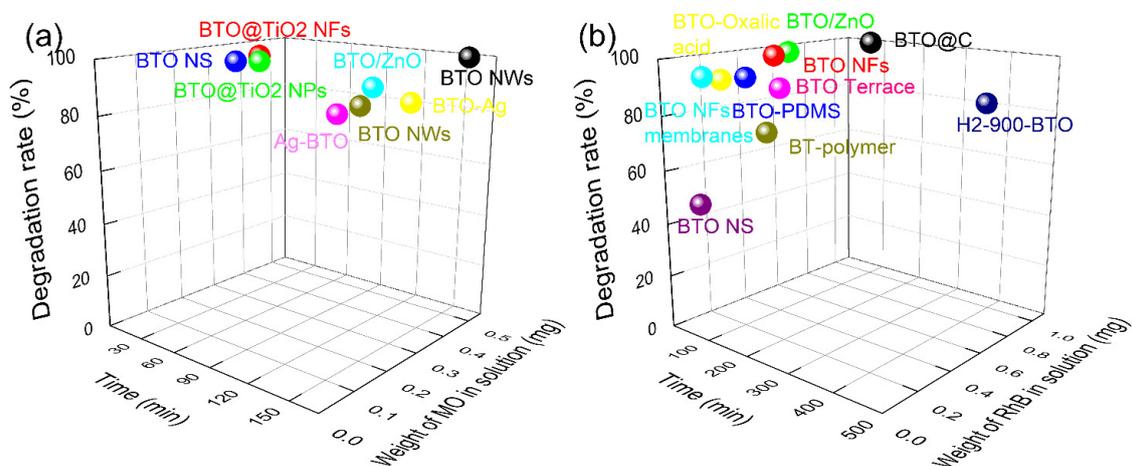


Figure 2. (a) Performance comparison of MO degradation. (b) performance comparison of RhB degradation. The data in this figure is sourced from Table 1.

Table 2. Degradation efficiencies and reaction conditions of BTO&US for different organic pollutants.

Number	System	Target	Initial Target Concentration (mg/L)	Volume (mL)	Frequency (kHz)	US Power (W)	Time (min)	Degradation Efficiency (%)	References
1	BTO-Ag	MO	5	100	40	120	120	~81	[9]
2	BTO	TMP	10	-	40	100	30	41	[10]
3	BTO NS	MB	2	50	35	180	40	~91	[12]
4	BTO NS	BPA	2	50	35	180	40	~61	[12]
5	BTO NS	TH	2	50	35	180	40	~55	[12]
6	BTO NS	phenol	2	50	35	180	40	~63	[12]
7	BTO NS	MO	2	50	35	180	40	~97	[12]
8	BTO NS	RhB	2	50	35	180	40	~47	[12]
9	BTO-PDMS	RhB	5	40	40	400	120	94	[15]
10	BTO-180+PMS	AMX	5	50	40	300	60	~80	[16]
11	BTO-180	AMX	5	50	40	300	60	~20	[16]
12	BTO NPs	IC	10	~50	40	80	35	90	[43]
13	BTO NWs	MO	5	100	40	180	80	~77	[44]
14	BTO NWs	MO	5	100	40	120	160	~100	[45]
15	Ag-BTO *	MO	10	50	24	120	120	~83	[46]
16	BTO@TiO ₂ NFs	MO	5	50	40	180	60	~100	[48]
17	BTO@TiO ₂ NPs	MO	5	50	40	180	60	~98	[48]
18	BTO Terrace	RhB	5	50	40	360	180	91	[49]
19	1mAg-BTO *	RhB	0.01 mM	50	-	-	75	83	[50]
20	BT-polymer	RhB	~5	10	40	70	240	80~90	[51]
21	BTO@C-0.01M *	RhB	10	100	40	120	100	~100	[52]
22	BTO NFs	RhB	7.5	50	40	-	110	~100	[53]
23	BTO/ZnO **	RhB	5	100	-	120	90	~100	[54]
24	BTO/ZnO **	MO	5	100	-	120	90	>85	[54]
25	BTO/ZnO **	LEVO	5	100	-	120	90	>85	[54]
26	BTO/ZnO **	MB	5	100	-	120	90	>85	[54]
27	BTO NFs membranes	TH	5	~20	40	240	45	82	[55]
28	BTO NFs membranes	MB	5	~20	40	240	60	96	[55]
29	BTO NFs membranes	RhB	5	~20	40	240	60	94	[55]
30	AgI/Ag ₃ PO ₄ /BTO	NTP	5	50	40	200	10	20	[56]
31	BTO-Oxalic acid	TH	5	-	40	100	60	~80	[57]
32	BTO-Oxalic acid	RhB	5	50	40	100	30	91	[57]
33	H ₂ -900-BTO	BPA	15	50	20	-	180	97	[58]
34	H ₂ -900-BTO	RhB	15	50	20	-	450	86	[58]
35	BTO powder	DS	-	-	40	70	150	78	[59]
36	BTO powder	CIP	-	-	40	70	150	85	[59]
37	BTO powder	MB	5	-	40	70	150	92	[59]
38	FTO/BTO/AgNPs	MB	5	75	24	-	180	90	[60]
39	FTO/BTO/Ag NPs	CIP	5	-	24	-	180	68	[60]
40	HPVDF/BTO-OV	BPA	15	20	40	300	60	98	[61]
41	BTO	SMX	5	20	45	300	15	~68	[62]
42	BTO NW+PS	IBP	6	25	40	110	60	~100	[63]
43	BTO NW	IBP	6	25	40	110	60	~80	[63]
44	t-BTO	4-CP	25	25	40	110	120	~71	[64]
45	t-BTO	3-CP	25	25	40	110	120	~58	[64]
46	t-BTO	2-CP	25	25	40	110	120	~51	[64]

*, With Xe lamp 300W; **, With Hg lamp 300W; NFs: nanofibers; PDMS: polydimethylsiloxane; NS: nanosheets; NWs: nanowires; NPs: nanoparticles; FTO: fluorine-doped tin oxide; Ovs: oxygen vacancies; HPVDF: hydrophilic porous polyvinylidene fluoride; PS: persulfate; PMS: Peroxymonosulfate; t-BTO: tetragonal BaTiO₃.

2.3.2. Application in Electrochemistry

After exploring the electrochemical performance of BTO&US, researchers have gained valuable insights that significantly expand its potential applications. The research on electrochemical batteries and electrochemical performance has laid a solid foundation for its application in clean energy and environmental remediation. It is innovative that ultrasound can continuously update the piezoelectric charge on the surface of BTO and trigger micro pseudo-electrochemical reactions. This approach transforms the entire system into micro pseudo-electronic cells, enabling in-situ synthesis of polyaniline on the surface [65]. In addition, the heterojunction of BTO and metal can significantly promote the separation and migration of electrons. This enhanced electron movement drives the electrochemical oxygen reduction reaction and hydrogen evolution reaction (HER) efficiently [66]. The ferroelectric heterostructure of BTO@MoSe₂ nanosheets can also be used in HER, and the highest record was achieved under piezoelectric catalysis at that time [67]. BTO nanosheets doped with Li and La can enhance the

rate of HER by 4.6 and 3.9 times, respectively [7]. Finally, ZnS/In₂S₃/BTO composites are instrumental in generating an H₂O₂ reaction when subjected to an external light source, which is also a new strategy in artificial photosynthesis [68].

2.3.3. Emerging Contaminants Degradation

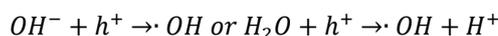
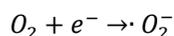
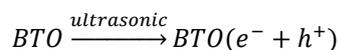
The degradation of emerging contaminants has emerged as a prominent research focus in recent years. These contaminants often exhibit distinctive characteristics such as biological toxicity, environmental persistence, and bioaccumulation. Common emerging contaminants can be roughly divided into persistent organic pollutants, endocrine-disrupting chemicals, antibiotics, microplastics, etc. These contaminants are typically present in small quantities but possess significant harmful potential, challenging their detection and effective degradation. Researchers actively seek efficient and secondary pollution-free degradation methods to tackle this issue. Some researchers are exploring the degradation of emerging contaminants using BTO&US. This method is considered environmentally friendly with low energy consumption, offering promising avenues for removing emerging contaminants. It has been demonstrated that BTO&US exhibits the capability to degrade various emerging contaminants such as ciprofloxacin (CIP), diclofenac, 4-chlorophenol, sulfamethoxazole (SMX), and BPA [58–60,62,64]. In addition, a wider range of free radicals can be generated when combining BTO with easily activated materials like PS and peroxymonosulfate (PMS). When coupled with PS, BTO has shown efficiency in degrading ibuprofen (IBP) [63]. When combined with PMS, BTO has effectively degraded amoxicillin (AMX) [63]. When BTO incorporates surface functional groups, the degradation ability of TH can also be obtained [57]. At the same time, some researchers have carried out application research utilizing BTO in environmental protection. Wan et al. embedded BTO nanocubes into a PVDF polymer membrane, enhancing its adsorption and activation capabilities for dissolved oxygen by adjusting the oxygen vacancy [61]. In this way, the material can effectively adsorb and degrade BPA, boasting a removal rate that was 14 times higher compared to a pure PVDF membrane. Moreover, they stick the material on the impeller, which also effectively removes BPA at 900rpm, providing the experience of low-frequency water purification.

3. Summary and Discussion of Underlying Mechanisms

The application of BTO&US materials is distributed in different fields. According to the mechanism research of various researchers in different applications, the mechanism of BTO&US can generally be categorized into three distinct stages. In the initial stage (depicted in Figure 3-1#), BTO absorbs ultrasonic vibrations. Numerous researchers have substantiated this phenomenon through theoretical calculations, notably finite element calculations. Furthermore, several characterizations, including Piezo-response force microscopy, X-ray photoelectron spectroscopy, and zeta potential analyzer, have been employed to investigate it. These techniques are primarily utilized to observe alterations in the piezoelectric potential and electron density of BTO before and after exposure to ultrasound. When subjected to ultrasonic assistance, BTO responds to the changes in pressure. This response manifests internally, affecting the material's piezoelectric polarization and changing the surface piezoelectric potential. From the perspective of the energy band, the band gap width of BTO and its band potential (in comparison to standard hydrogen electrodes) are not conducive to generating free radicals. Upon exposure to ultrasonic waves, the energy band will be tilted to mitigate the adverse effects during the generation of free radicals. From the perspective of electrons, electrons and holes are more easily separated after BTO absorbs ultrasound. Therefore, the density of charge carriers is increased, which makes the redox reaction near the valence and conduction bands more likely to occur. Looking at the crystallography perspective, some researchers consider that under the influence of ultrasound, Ti⁴⁺ within BTO will undergo an eccentric shift, which indirectly changes its internal electric field. Moreover, the presence of oxygen vacancies is a significant factor of interest because BTO prepared by calcination has a certain amount of oxygen vacancies. Some researchers also indirectly verify the role of charge carriers in catalysis through oxygen vacancies. In addition, investigating the changes in electron density caused by electron transfer is also a promising research direction. Density Functional Theory calculations and establishing a differential charge density model are practical tools for this research. At this stage, there is a consensus in the scientific community that BTO undergoes modifications under ultrasound, making it prone to produce active substances.

In the second stage (depicted in Figure 3-2#), active substances form around BTO. Researchers commonly employ electronic spin resonance or design experiments to capture free radicals to detect the active substances produced in the process of piezoelectric catalysis. Researchers generally agree that these active substances primarily manifest as free radicals, which may be ·O₂⁻, ¹O₂, ·OH, holes, etc. Several factors contribute to the production of these active substances. Some researchers propose that, under the initial conditions, the potential of BTO itself meets the criteria for free radical formation but lacks free charge. When under ultrasonic conditions,

free charge exists inside the material, enabling the generation of free radicals [69]. Additionally, some researchers posit that BTO may release charge on the surface under periodic pressure induced by ultrasound and initiate two opposing redox reactions to form free radicals. Undeniably, a distinct redox reaction occurs in this process, inevitably involving electron transfer. The movement of these electrons is related to ultrasound. In the preceding stages, electrons and holes are created in BTO and transferred to the water, which reacts with substances in the water to generate free radicals, as demonstrated by the ensuing chemical reactions:



In the biological field, these active substances may be related to hormones or ions. The alterations in the electric field and electric potential of BTO under the influence of ultrasound stimulate the organism, consequently impacting biological processes like protein synthesis, hormone secretion, and ion transfer.

In the third stage (depicted in Figure 3-3#), active substances are applied in various fields. In the field of organic decomposition or antimicrobial therapy, this aspect is typically straightforward. Reactive oxygen species exhibit potent oxidation capabilities, allowing them to complete their mission through redox reactions. In terms of organic decomposition, free radicals will trigger a series of chain decomposition reactions to decompose large organic compounds into smaller molecules. Gas chromatography or liquid chromatography combined with mass spectrometry can analyze the final products. In terms of biology, bacteria, and cells are composed of various organic compounds, and similar redox reactions will occur to achieve antimicrobial therapy results. It is also possible that the growth or apoptosis of bacteria or cells can be caused by regulating hormones or controlling ion permeability in cells. Compared to mere inhibition, the latter approach may lead to cell death. Flow cytometry and electron microscopy areas are used to distinguish them. These techniques can determine whether cells or bacteria are decomposed or apoptotic by assessing the proportion of living and dead cells. Moreover, a scanning electron microscope can be used to observe cell images, and some methods can be used to detect protein leakage, changes in malondialdehyde concentration, membrane permeability, oxidative stress degree, etc. These analytical methods aid in understanding the causes of cell injury or death.

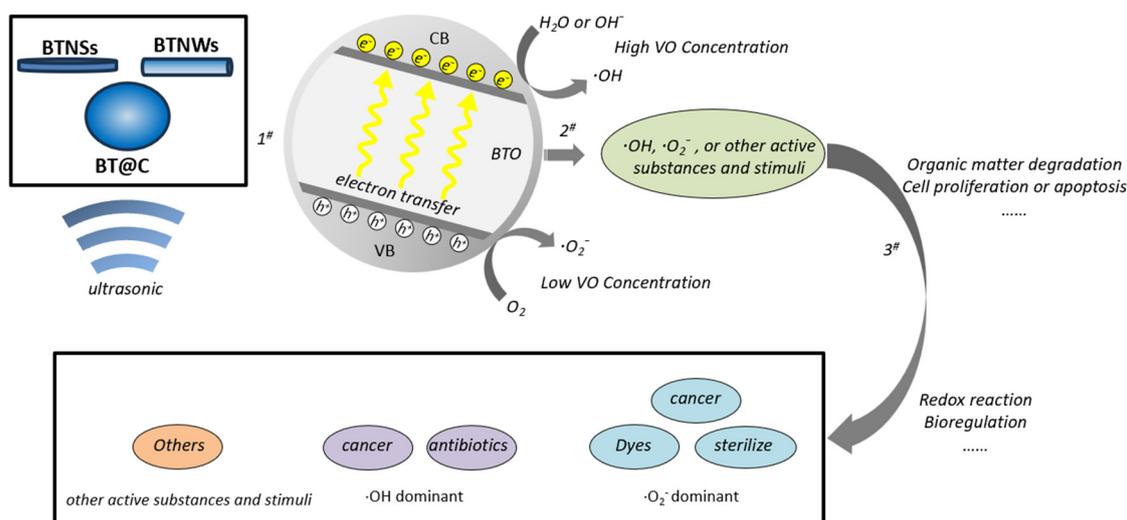


Figure 3. Mechanism summary of BTO&US.

4. Perspective of Future Studies

It can be found that BTO&US presents numerous advantages in applications, forming the foundation for its extensive application. Firstly, its wireless nature liberates it from the shackles of connecting wires, enabling the non-contact application of vibration to BTO. Secondly, the power and frequency of the ultrasonic generator can be adjusted in real-time, enabling indirect modification of the piezoelectric effect of BTO. This can achieve the

corresponding effect according to individual requirements. Thirdly, the free radicals produced by piezoelectric catalysis are strong oxidants and have universality. They can directly break down various pollutants into harmless small molecular substances. Fourthly, BTO&US acts as a catalyst and does not participate in the reaction process. Compared with physical methods like filtration or biological methods like the activated sludge method, no contaminant-laden filter media is challenging to treat. Additionally, the process eliminates the need for sedimentation and sludge treatment. This avoids secondary pollution and is also a green method. The improvement of barium titanate can promote the material's performance, roughly divided into the following aspects: (1) Morphology and crystal structure. It can influence the material's electron transfer efficiency, specific surface area, and vibration absorption ability. For example, nanowire morphology can better absorb ultrasound due to its structure. (2) Doping or loading. This can adjust the energy band structure of materials and leverage their interactions, which is more conducive to separating electron-hole pairs. (3) Synergies. By combining BTO with auxiliary materials, the piezoelectric effect of BTO can stimulate the activity of auxiliary materials, generating a diverse array of free radicals and enhancing the overall performance.

4.1. Research Direction

The application mechanism of BTO&US is relatively straightforward, yet some interesting points still deserve in-depth study. For instance, (1) The study between crystal structure and electrons under the influence of ultrasound. The crystal structure affects the piezoelectric catalytic ability of BTO. How this affects the charge distribution of materials is worth discussing. (2) The alteration in surface electron density before and after ultrasound can be indirectly demonstrated through changes in ionic valence before and after the material receives ultrasonic vibration. (3) The influence of the medium on the catalytic activity. Current research predominantly employs water as the solvent, yet exploration with other organic solvents remains limited. Diversifying solvents may broaden the spectrum of generated free radicals. (4) The process of interacting with microorganisms. For example, bacteria may undergo mortality during sterilization due to physical destruction, oxidation-reduction reactions, or hormone regulation. The mechanism of this process can be explored. (5) The study on the interaction between organic matter and materials. Whether the organic dyes will be adsorbed on the surface of BTO to react with the active substance and the principle of this reaction remains unexplored. Addressing these inquiries will propel BTO&US research into a more comprehensive and in-depth domain.

4.2. Application Direction

Emerging contaminants have been a hot research topic and an emerging global problem in recent years. Traditional wastewater treatment approaches, including biological methods like activated sludge and biofilm processes, alongside physical methods like activated carbon adsorption and membrane filtration, as well as chemical methods such as chlorination, permanganate oxidation, and ultraviolet irradiation, often fall short in eliminating these emerging contaminants [70]. The Advanced Oxidation Process with free radicals as the primary active substance provides a basis for developing new treatment methods. Free radicals exhibit several advantages attributed to their strong oxidizing properties [71]. For example, (1) they can eliminate organic pollutants that pose challenges for biodegradation; (2) they can degrade chemically stable organic pollutants; (3) they can reduce trace organic pollutants under mild conditions; (4) fewer by-products generated after degradation. Advanced Oxidation Processes, including photocatalysis and the Fenton process, have been successfully applied in this field. Photocatalysis, for instance, is capable of degrading over 30 emerging contaminants [70], such as microcystin algae toxins [72,73], diclofenac [74], IBP [75], tetracycline [76], and caffeine [77]. Besides, more than 20 types of emerging contaminants have been degraded using the Fenton process [78,79], *Microcystis aeruginosa* (*M. aeruginosa*) and Microcystin-LR can also be reduced under the action of free radicals [80]. The Fenton process primarily relies on peroxide and ferrous ions to generate hydroxyl radicals under acidic conditions [81]. This illustrates that techniques generating free radicals can be explored to degrade emerging contaminants. Thus, by analyzing its mechanism, it becomes evident that BTO&US holds significant potential for application in the emerging contaminants field [71,82].

Hence, the degradation of emerging contaminants is a prospective research avenue for BTO&US. Several potential directions are worth exploring: (1) As for microplastics, limited research has been conducted on laboratory-scale exploration. BTO&US lacks relevant research in this field for the time being and can endeavor to investigate polystyrene degradation [83]. (2) As for microcystin, eutrophication remains a prevalent water pollution issue, drawing increasing attention to the microcystin contamination associated with it. Similarly, there is a lack of relevant articles on applying BTO&US in this area. Microcystin-LR can be used as the object of the experiment to investigate its degradation potential [72,80]. (3) As for hormone pollutants, researchers have proven

that BPA can be degraded to expand more pollutants. If BTO&US is considered for practical application, it is necessary to consider some factors that affect it, such as pH, coexisting ions, and temperature, because the real environmental conditions for these pollutants are complex. These factors may reduce the activity of materials. Hence, in practical applications, it is important to consider water pretreatment measures (e.g., coagulation, sedimentation, ion exchange, water softening) to optimize the effectiveness of this method. In addition, this method may also have many daily application scenarios. For example, BTO can be used as a coating or additive material in household ultrasonic toothwashers, dishwashers, and cleaning instruments.

5. Conclusions

This review offers an extensive overview of the evolving landscape in BTO&US research spanning various domains. The method boasts numerous merits, including wireless control, universality, precise control, and biosafety. Its applications have demonstrated significant potential across diverse realms, such as cancer treatment, auxiliary medical materials, antimicrobial therapy, cell control, organic degradation, and electrochemistry. The application mechanism is also summarized and can be delineated into three stages: vibration absorption, active substance generation, and application in various scenarios. This paper focuses on elucidating the mechanism and exploring future research directions for this method. The research topics of mechanisms about crystal structure, electron density, medium, and interaction are also discussed, with a particular emphasis on the degradation of emerging contaminants as a key research focus for this method in the future. Furthermore, the paper presents research prospects related to microplastics, microcystin, and hormone pollutants. The comprehensive analysis of BTO&US can provide valuable guidance for exploring novel fields and mechanisms, aiming to unlock the potential of barium titanate materials further and promote wider adoption of piezoelectric ceramics applications.

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