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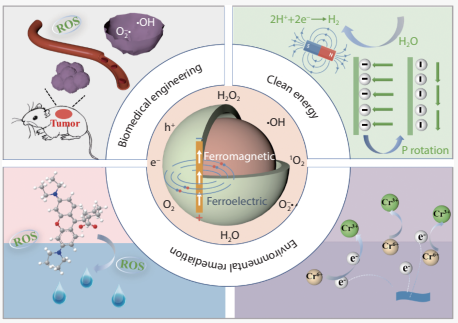
REVIEW ARTICLE

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Magnetic field driven catalysis of multiferroic magnetoelectric nanocomposites

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Magnetic field as a booster for catalytic reactions has been widely studied in the past few decades. Recently, multiferroic materials with intriguing magnetoelectric coupling effects have been emerging as a new type of catalyst, providing a unique opportunity for magnetically-driven catalytic reactions in a variety of fields, including clean energy, environmental and biomedical applications. In this review, we describe this entirely new catalysis phenomenon observed in multiferroic magnetoelectric composite materials, aiming at giving an in-depth understanding of magnetically-driven catalysis processes based on the direct magnetoelectric-catalytic effect. Moreover, the latest progress in catalytic applications of magnetoelectric nanocomposite nanomaterials is comprehensively summarized. Finally, the challenges and future perspectives for the design and application of high-efficient magneto-multiferroic catalysts are discussed.



With the increasing worldwide concern of urgent energy crisis and environmental pollution issue, the development of efficient catalysis systems and strategies that can initiate or accelerate target chemical reactions is of great importance. In general, introducing an external field (e.g., thermal, electric, magnetic, and mechanical) would lead to improved efficiency of an existing catalytic system.[1-6] Among reported routes, the employment of a magnetic field has been considered as an effective strategy, and significant advances have been achieved in different catalysis fields, including photocatalysis,[7-11] electrocatalysis,[11-17] and thermocatalysis,[18] leading to significantly enhanced performance and allowing for wireless control of catalytic reactions.[19, 20] It is concluded that magnetic fields can affect the spin polarization of magnetic materials, thereby accelerating the movement of free charge carriers and increasing the rate of electron transfer to the surface and generation of free radicals participating in catalytic reactions.[1, 7, 21] As well as being an invaluable variable, the magnetic field can be used as an effective controller of both the product and the rate of a chemical reaction process.[22] It should be noted that generally the magnetic field could not directly generate charges and internal electric field required for a catalytic reaction, and thus proper magnetic materials are crucial in magnetically-assisted catalysis processes.

Multiferroic materials, which exhibit ferroelectric and ferromagnetic characteristics simultaneously, have stimulated increasing research activities during the past few decades for both fundamental science and potential technological applications in next-generation multifunctional devices, such as non-volatile memory, spintronics, photovoltaics, and acousto-optic devices.[8, 23-26] In particular, the unique advantage of multiferroic materials is the coupling of magnetoelectric states, which enables new functionalities that neither of the individual states possesses.[27-29] Driven by the magnetic field, the direction change of electrical polarization will produce surface charges in multiferroic magnetoelectric materials,[30] which will trigger catalytic reactions. Therefore, multiferroic magnetoelectric nanocomposite materials are ideally suitable for inducing and manipulating of catalytic reactions solely by external magnetic fields. In 2019, a pioneering work reported by Mushtaq *et al*. firstly demonstrated that magnetoelectric $CoFe_2O_4$-$BiFeO_3$ nanocomposites are able to decompose organic contaminants by simply applying an AC magnetic field without the use of any sacrificial molecules or cocatalysts,[31] shedding light on the utilization of multiferroic magnetoelectric nanomaterials as magnetically-triggered

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catalysts. Later on, several studies have proved the general applicability of this novel catalysis effect by expanding their applications into different areas such as hydrogen production,[32] pollutant treatment,[33] and biomedical applications.[34, 35]

Different from previously published review articles on conventional magnetically controlled catalytic chemical conversions,[7-16] herein we focus on the emerging multiferroic magnetoelectric nanomaterials-based catalysis employing magnetic fields as the ultimate trigger. Begin with a brief introduction of multiferroic/magnetoelectric effect, a thorough state-of-the-art overview of magnetically-driven magnetoelectric-catalytic reactions and related applications, including clean energy (e.g., H_2 production), environment remediation (e.g., organic pollutant degradation and toxicant removal), and biomedical engineering (e.g., eliminating bacterial organisms), is presented. Finally, in view of the future development direction of magnetically-driven multiferroic magnetoelectric nanocomposite materials in catalysis, we put forward prospects and solutions for future challenges and perspectives.

Multiferroic magnetoelectric nanomaterials

According to the origin of magnetoelectric coupling, multiferroic materials where two or more primary ferroic orders coexist,[28, 36, 37] can be roughly classified into two categories: (i) single-phase multiferroics and (ii) heterogeneous multiphase composites consisting of individual magnetostrictive and ferroelectric/piezoelectric components. The origin of multiferroicity is mainly related to magnetoelectric coexistence and magnetoelectric coupling effects.[38-40] The magnetoelectric coexistence mechanism is primarily dominated by charge ordering, lone pair electrons, geometry and spin effects, while the magnetoelectric coupling mechanism is originated from exchange coupling, strain surface, domain and domain wall effects.[41-43]

In the past few decades, a great number of single-phase multiferroic materials have been developed such as $BiFeO_3$, $TbMnO_3$, and $LuFeO_3$.[8, 44-48] Among them, $BiFeO_3$ is the most widely studied multiferroic materials with robust ferroelectric and magnetic orderings at room temperature.[49] The ferroelectric polarization in $BiFeO_3$ is mainly induced by the ordered $6s^2$ lone pair electrons of Bi, and its antiferromagnetism comes from the 3d electrons of Fe.[50] Nevertheless, many limitations have been imposed on single-phase multiferroic magnetoelectric materials because of the d-electrons of constituent metal ions (e.g., Fe^{2+} and Fe^{3+}) that are essential for magnetism (partially occupied d-orbitals) but reduce ferroelectric order (empty d-orbitals). In contrast, heterogeneous multiphase materials composed of ferromagnetic and ferroelectric phases can induce a piezo-magnetoelectric coupling effect between the electric and magnetic sub-systems at the interface. Up till now, well-designed composites have been developed as an intriguing alternative option for multiferroic materials,[29, 51] as exemplified by $CoFe_2O_4/BiFeO_3$,[52, 53] $Pb(Zr,Ti)O_3/CoFe_2O_4$,[54, 55] $Pb(Zr,Ti)O_3/Fe_{0.7}Ga_{0.3}$,[56] and $La_{0.7}Sr_{0.3}MnO_3$/P(VDF-TrFE),[57] with strong magnetoelectric coupling that are orders of magnitude higher than in single phase. With the increasing demands for further miniaturization of electronic devices, the design and fabrication of nanostructured multiferroic composites is of great importance.[58] For instance, Eerenstein *et al*.[59] demonstrated a predominant electrically-induced magnetic change in an epitaxial heterostructure consisting of ferromagnetic $La_{0.7}Sr_{0.3}MnO_3$ film grown on ferroelectric $BaTiO_3$ substrate, in which the switching of $BaTiO_3$ domains alters the local strain, magnetic anisotropy and thus the magnetization in the $La_{0.7}Sr_{0.3}MnO_3$. In addition, Sreenivasulu *et al*.[60] reported core-shell structured multiferroic nanocomposites by functionalizing $BaTiO_3$ and $NiFe_2O_4$ nanoparticles (10 to 100 nm) with complementary coupling groups, demonstrating strong strain mediated magneto-electric coupling under static magnetic field induced variations with the permittivity over 16 to 18 GHz.

Magnetically driven catalysis of magnetoelectric nanocomposites

In previous studies, multiferroic nanomaterials have been already used in various catalysis areas including photocatalysts, electrocatalysis, and piezocatalysts, such as $BiFeO_3$,[61-65] Bi_2FeCrO_6,[66, 67] $TbFeO_3$,[68] $KBiFe_2O_5$,[69] and $Bi_5CoTi_3O_{15}$,[70] owing to their unique advantages of suitable narrow band gap and predominant ferroelectric/piezoelectric response. Effective manipulating of charge transport and catalytic performance by ferroelectric polarization induced electric filed have been achieved in these multiferroic nanostructures.[71, 72]

Recent works demonstrated an intriguing direct strain-mediated magnetoelectric-catalytic effect in multiferroic nanocomposites, which allows magnetic field stimulation of chemical reactions at the polarized surface, providing an entirely new catalytic mechanism.[32, 35] In this process, once an AC magnetic field (H_{ac}) is applied to the magnetoelectric composite materials, the induced strain in the magnetostrictive phase is transmitted to the ferroelectric (i.e., piezoelectric) phase, resulting in a change in electrical polarization. Therefore, multiferroic magnetoelectric composite materials are particularly suitable for initiating and controlling catalytic reactions driven by magnetic fields, as their electrical polarization and surface charges can be magnetically altered. Based on the direct "magnetoelectric-catalytic" effect, there has been increasing interest in the application of multiferroic materials to trigger different catalytic reactions. It is confirmed that many reactive oxygen species (ROS) (e.g., $^1O_2$, •OH, $•O_2^-$, and H_2O_2) could be stimulated on the surface of the multiferroic magnetoelectric catalysts by solely applying a magnetic field.[34, 35] As shown in Fig. 1, the representative applications of magnetically-driven multiferroic materials that have been reported to date in the field of catalysis are presented, involving environmental remediation, clean energy, biomedical engineering, etc. In comparison with other input energy sources such as light and ultrasound, the large penetration depth of magnetic fields allows full activation of catalysts in aqueous media.

Environmental remediation

Organic contaminants, the main constituents of water pollution, are difficult to degrade by conventional processes and thus cause serious environmental issues. Up till now, a great

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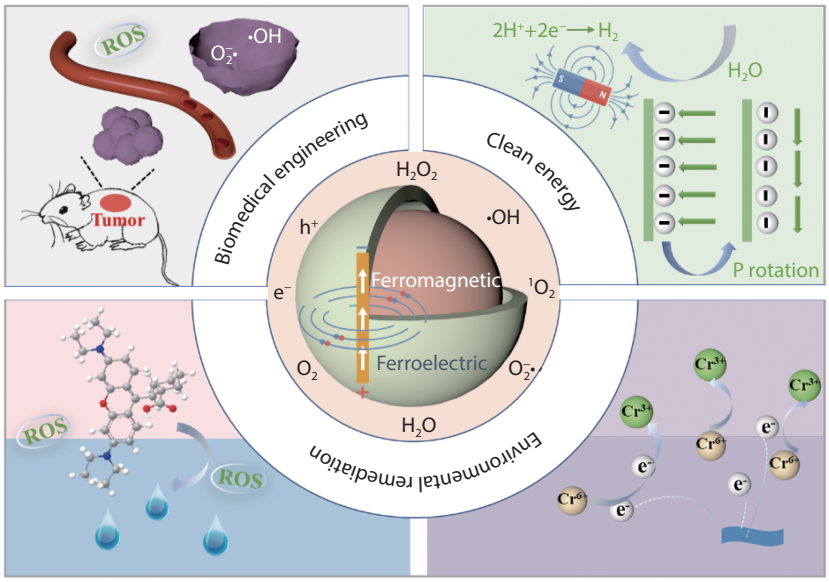


Fig. 1 Representative applications of multiferroic magnetoelectric nanocomposites in catalysis driven by magnetic fields.

number of strategies have been proposed for removing organic pollutants in water, including advanced oxidation processes (AOPs),[73] photocatalysis,[74] and piezocatalysis, etc.[5, 75] The magnetoelectric-catalytic effect provides an attractive method for the degradation of organic pollutants upon magnetic fields. Mushtaq *et al*.[31] firstly utilized core-shell structured $CoFe_2O_4/BiFeO_3$ (CFO/BFO) nanoparticles as catalyst for removing organic pollutants (RhB as the model dye), reaching a high efficiency of 97% in only 50 min under an AC magnetic field (15 mT and 1 kHz), without using any sacrificial molecules or co-catalysts. Under the magnetic field, electrons and holes are formed on the surface and reacted with the surrounding oxygen and water to form $•O_2^-$ and •OH, respectively, which are responsible for the magnetoelectrically induced catalytic degradation of organic compounds. This pioneering work demonstrates the promising potential of multiferroic materials in magnetically driven catalysis for environmental remediations.

In addition, the conversion of highly toxic valence states to less toxic valence states by catalytic redox reactions (e.g., Cr^{6+} to Cr^{3+}[76, 77]) is of great importance for the treatment of heavy metal polluted water environment. Although photocatalytic reduction of heavy metals has been widely investigated, it still suffers from the shortcomings of low efficiency in the treatment of turbid and colored polluted water due to the shading effect of light.[78] Recently, Mushtaq *et al*.[33] reported core-shell magnetoelectric CFO/BFO nano-octahedral particles for Cr^{6+} reduction, presenting a new approach for removing toxic metal ions in water.

Clean energy

Producing H_2 from water has been widely recognized as a clean and low-cost strategy with promising prospect in the future. Hydrogen evolution reaction (HER) through water splitting has been successfully achieved driven by electrical, solar, mechanical, and thermal energy, but these technologies still suffer from low efficiency or high cost.[79-82] Therefore, it is necessary to develop alternative approaches for water splitting. In 2022, Kim *et al*. firstly utilized multiferroic CFO/BFO core-shell composite catalysts for HER upon an AC magnetic field, reaching a H_2 production rate of 28.7 μmol g^{-1} in 6 h (Fig. 2a).[32] The hydrogen evolution rate enhanced with increasing the intensity and frequency of the applied magnetic field (Fig. 2b and 2c), which could be attributed to the larger induced magnetostrictive strain. After dispersing in $AgNO_3$ solution, the formation of Ag nanoparticles on the surface of the CFO/BFO nanoparticles clearly verify the magnetically generated electrons by the magnetoelectric effect. Based on the experimental results and density functional calculations, the mechanism could be attributed to the polarization switching caused by the strain gradient via magnetostriction, local polarization inversion by the motion of the domain wall, and the parallel rotation of the ferroelectric polarization that prevents the charge compensation (Fig. 2d).

Biomedical applications

The magnetic fields have been proven to be highly biocompatible with human cells (e.g., osteoblasts, neuronal cells, and mouse models).[35, 83, 84] For example, magnetic resonance imaging (MRI) is widely used to map the brain. Moreover, magnetic fields are external stimulus with superior tissue penetration depth but cause minimal damage to normal tissues than other sources such as ultrasound, infrared light, and X-ray irradiation. These advantages indicate promising prospects of magnetoelectric effects for biomedical applications.

In 2021, Ge *et al*. firstly reported magnetoelectric-catalytic effect induced tumor therapy.[35] Within 8 h, the uptake of cancer cells reached as high as 96.4% using CFO/BFO multiferroic composites under magnetic field. As shown in Fig. 3a, by monitoring the degradation of 1,3-diphenylisobenzofuran, it was confirmed that the magneto-piezoelectric-catalyzed ROS ($•O_2^-$ and •OH) were generated over the CFO/BFO nanocomposites due to the micro-strain and electric field induced by the magnetic field, which is supported by the COMSOL simulations. This work demonstrates a simple and effective magneto-triggered catalytic modality for cancer therapy based on magnetoelectric nanocomposite materials, which

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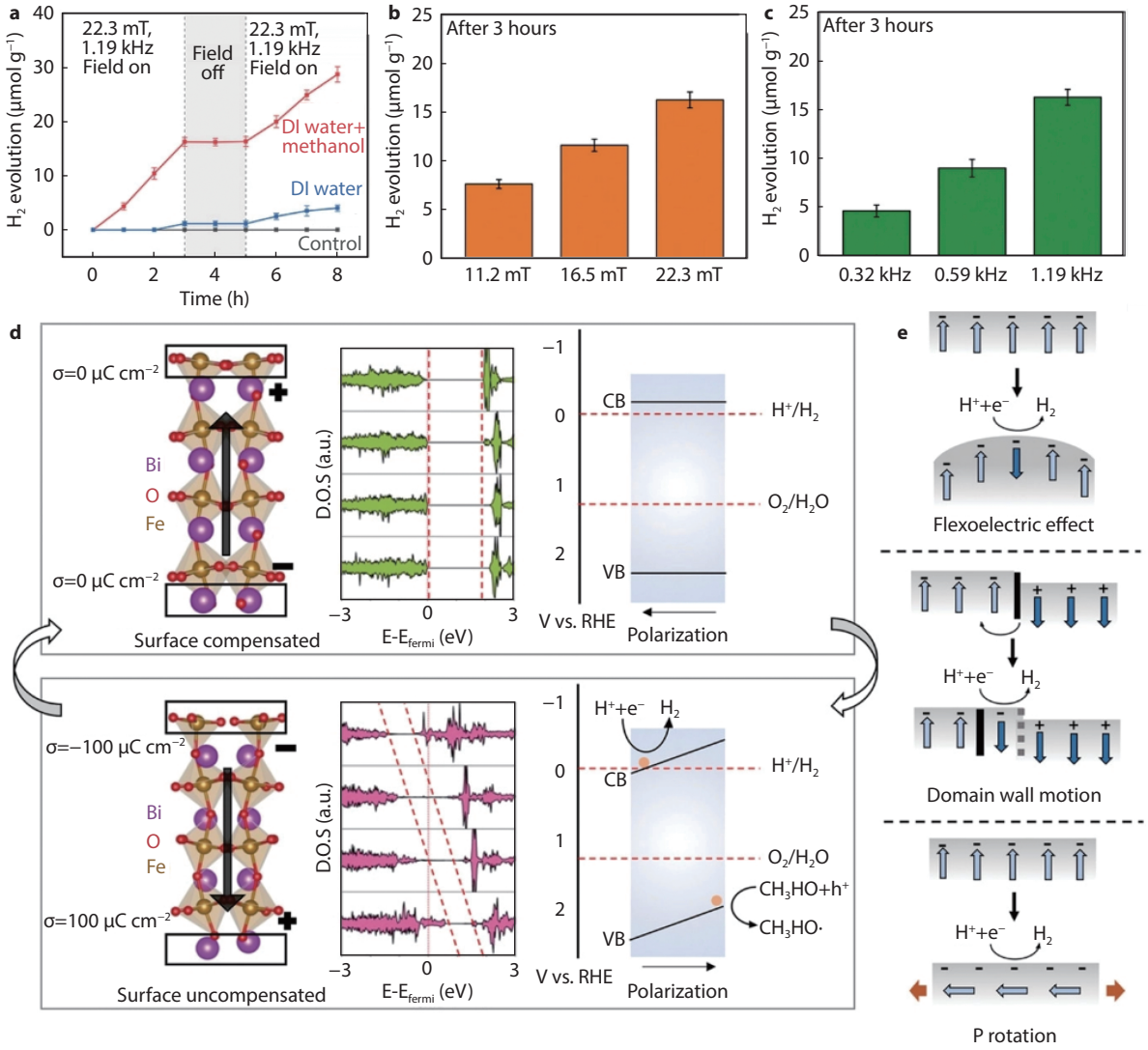


Fig. 2 **a** Hydrogen evolution curves of multiferroic CFO/BFO composites under an AC magnetic field. Influence of **b** frequency and **c** intensity of applied magnetic field on hydrogen evolution. **d** Calculated BFO (001) slabs and schematic diagrams of the band bending caused by the reversal of the polarization for up- or down-polarization states. **e** Proposed mechanisms for the polarization reversal by magnetoelectric coupling in the core–shell CFO/BFO nanoparticles for driving hydrogen production reaction.[32] Copyright 2022, Wiley-VCH.

holds promising prospects for clinic translation in the future without using toxic chemotherapeutic drugs. In addition, Deng *et al*. developed a novel nanozyme based on magnetoelectric CFO/BFO nanocomposites, which can generate abundant surface charges under an alternating magnetic field and establish controllable electron transport between the nanozyme-eel and bacteria, thus effectively eradicating bacterial biofilms.[85] Moreover, Dong *et al*. designed and fabricated a novel 3D printed helical micro-swimmer combining biodegradable soft hydrogel as the substrate and magnetoelectric CFO/BFO core-shell nanocomposites (Fig. 3b), which allows better delivery of catalysts in the body and induces cell differentiation via transient charge changes,[83] inspiring new routes for targeted cell therapies for traumatic injuries and diseases. Very recently, Jang *et al*. proposed low-frequency magnetic field-induced dissociation of toxic A aggregates, a major pathological feature of Alzheimer's disease (AD), using CFO/BFO nanoparticles (Fig. 3c), unveiling the promising potential of magnetoelectric-catalytic materials for AD treatment.[34]

Multi-stimuli activated magnetoelectric-catalysts

Multiferroic/magnetoelectric materials concurrently possess multifunctionalities such as ferro-/piezo-/pyroelectric and photoelectric properties. Consequently, the coupling of these characteristics with magnetoelectric-catalytic effect allow these materials with intriguing prospects for simultaneously harvesting diversiform energy sources such as light, mechanical, and waste heat energy, leading to enhanced performance and better adaptation to complex systems for practical applications. The combination of various input energy sources is expected to be favorable for overcoming the limitation of applying solely magnetic field.

Dhanalakshmi *et al*.[86] synthesized ternary nanocompos-

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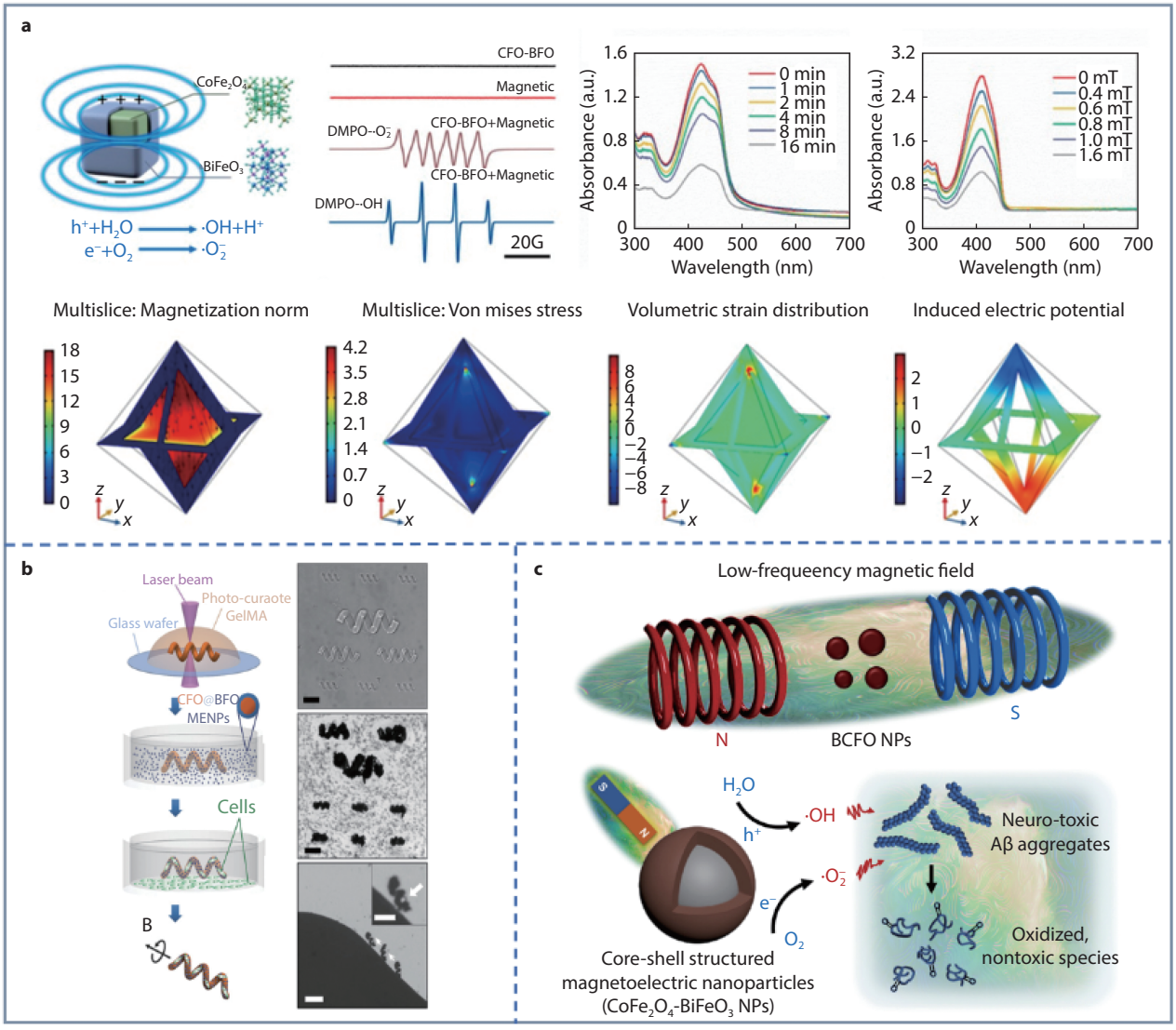


Fig. 3 **a** Mechanism illustration/experimental probing and COMSOL simulations for the magnetostrictive-piezoelectric catalytic effect under magnetic field over CFO/BFO nanocomposites.[35] Copyright 2021, American Chemical Society. **b** Schematic illustration of the fabrication process of biodegradable soft helical microswimmers based on CFO/BFO nanocomposites and the corresponding optical image.[83] Copyright 2020, Wiley-VCH. **c** Schematic illustration of magnetoelectric-catalytic dissociation of Alzheimer's Aβ aggregate structure using CFO/BFO nanoparticles under a low-frequency magnetic field.[34] Copyright 2022, Science.

ites consisting of $BiFeO_3/CoFe_2O_4/Co_3O_4$ with p-n-p heterojunctions for photocatalytic reduction of Cr(VI) under visible light irradiation. When 0.4 T external magnetic field was applied, the photocatalytic reduction rate increased by about 5 times, which is attributed to the effective electron-hole separation and transfer facilitated by the magnetic field and the interfaces. In addition, Mushtaq *et al*.[87] fabricated CFO/BFO nanocomposite photocatalysts with octahedral, cubic and nanowire morphologies, which allow simultaneous harvesting of multiple energy sources, including solar light, mechanical vibrations and magnetic fields. The rate of degradation for organic pollutants is significantly increased compared to the single-energy source stimulation. Very recently, Dong *et al*. designed and fabricated a magnetoelectric-photocatalytic heterostructure system composing of ZnO nanorods array grown on Metglas (Fig. 4).[88] The magnetoelectric coupling in magnetostrictive phase (Metglas ribbon) under the applied magnetic field causes an electric potential in the piezoelectric ZnO nanorods, resulting in a band shifting of ZnO. This synergistic magnetoelectric-photocatalysis effect leads to a significantly enhanced degradation rate (~3 times) under both weak AC magnetic field (H_{ac} of 2 Oe) and visible light, as compared to that under only light radiation. The generated local surface potential and built-in field via magnetoelectric coupling promotes the separation of photogenerated free charge carriers, resulting in improved degradation activity of dyes and disinfection in polluted water.

Conclusions and perspectives

Magnetic field driven or promoted nanocatalytic reactions have attracted increasingly attention and achieved remarkable progress. Multiferroic magnetoelectric nanocomposites have been emerging as promising candidate catalysts based

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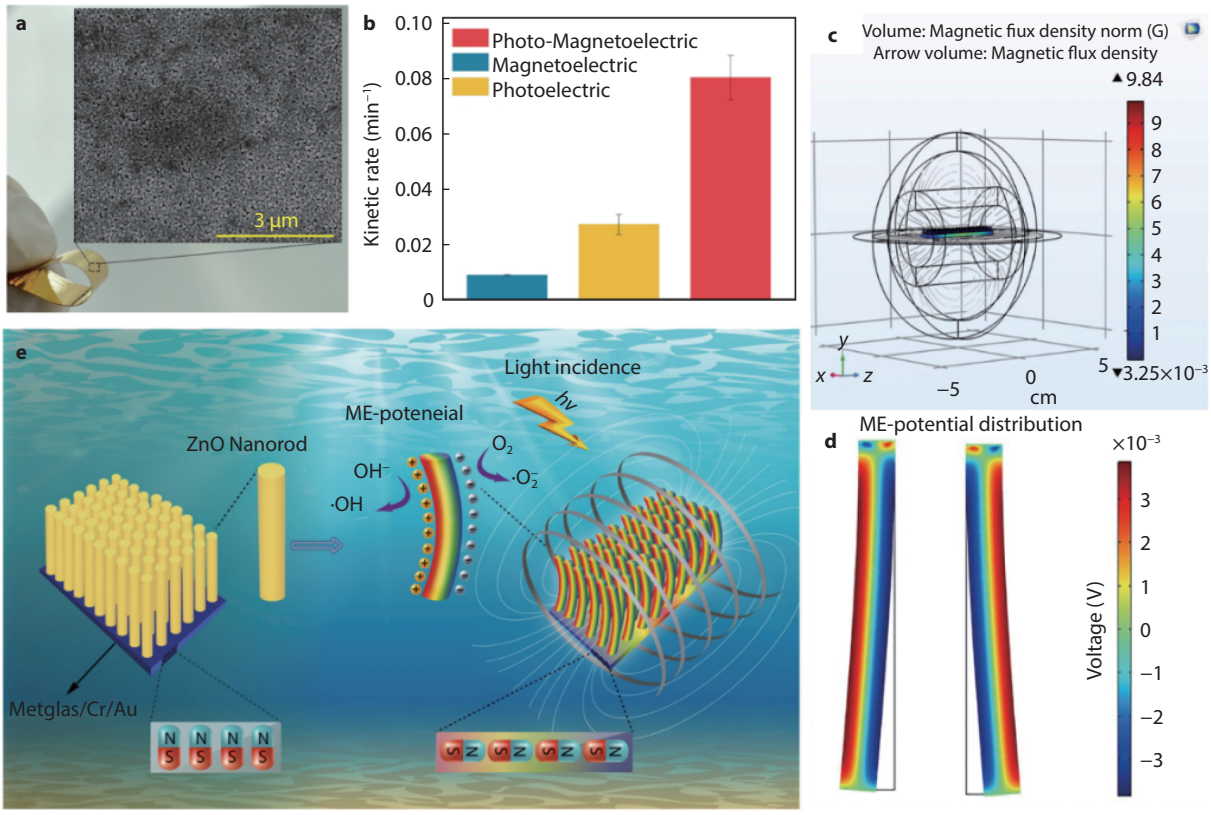


Fig. 4 **a** Digital and SEM image of ZnO nanorods array grown on a Metglas foil. **b** Kinetic rate for the degradation of indigo carmine under different conditions. **c** The magnetic flux density distribution in ZnO nanorods and Metglas under uniform magnetic field, and **d** the magnetoelectric potential distribution in a single ZnO nanorod under lateral deformation. **e** Schematic illustration of the magnetoelectric-photocatalytic mechanism for organic pollutant degradation process.[88] Copyright 2023, Elsevier.

on the direct magnetoelectric effect driven by solely magnetic field, opening up new avenues for magnetically induced catalytic reactions. In this review, we summarized the research progress of multiferroic nanocomposites in the field of magnetoelectric-catalysis towards diverse applications such as clean energy, environment remediation and biomedical engineering. At this moment, the development of this novel catalytic effect and the related applications are still at the early stage with many challenges and exciting opportunities. Herein we attempt to look at the current deficiencies and propose possible future development directions, aiming at promoting the leap-forward advance of this new research area.

So far, the investigated magnetically-driven multiferroic nanomaterials for catalysis are mainly focused on the well-studied CFO/BFO heterostructure system. The diversity of candidate magnetoelectric-catalysts needs to be further enriched, including other oxide-based nanocomposites[54-56, 89, 90] and polymer-based nanocomposites,[91-93] to promote the magnetoelectric coupling effect and improve the reaction kinetics by tuning the physical and chemical properties. Moreover, the emerging 2D materials with extraordinary physicochemical properties (e.g., high specific surface area, high charge mobility, and rich surface-active sites) hold great promise to serve as nanocatalysts for different catalytic applications. Thus, the development of 2D magnetoelectric nanocomposite catalysts, which are so far less addressed but definitely appealing, is highly desired.[94, 95] It is well known that the configuration of the magnetoelectric composites (e.g., 0-3 type particulate composites, 2-2 type laminate composites, and 1-3 type fiber/rod composites) are crucial for determining their coupling properties.[29] In this regard, advanced synthetic strategies are highly demanded for the rational engineering of magnetoelectric nanocomposites with desirable crystal facets, interface/surface structures, and configurations towards high-efficient catalysis systems to further advance this promising research field.

In addition, currently the magnetoelectric-catalysis mechanism remains unclear and thus both experimental and theoretical verifications are highly required to elucidate the underlying mechanism. It is highly important to establish the relationship among the structure of multiferroic magnetoelectric materials, the key parameters of the applied magnetic field (i.e., intensity and frequency), and the surface catalytic reactions. In previous works, the magnitude of magneto-induced compressive force and electric polarization field could not be measured precisely, making these studies lack of quantitative analysis. Therefore, well-designed in-situ experiment and theoretical calculations are of great significance to get a deeper understanding of the magnetoelectric-catalytic effect. Furthermore, it is necessary to elucidate the relationship between the electronic structures of the magnetoelectric composite and catalytic reaction kinetics (e.g., the energy level for thermodynamic reaction, the production and transfer of free charges, and the generation of free radicals on the

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surface) for specific reaction processes at the molecular/atomic level, to further pave the way for the design of efficient magnetoelectric-catalysts in the future.

Magnetic field driven catalysis over magnetoelectric composite materials is a rather new field, currently mainly focusing on the applications for water splitting, organic pollutant degradation and cancer therapy. Undoubtedly, these fascinating results in previous studies are anticipated to further stimulate and extend the research for other catalytic applications (e.g., selective growth, organic synthesis, and CO_2 reduction). Among all, biomedical engineering is the most likely to be implemented potential application, since the magnetic field is noninvasive or merely causes minimal damage to normal tissues, presenting enormous advantages. It is a highly interdisciplinary research field. First, it is crucial to develop proper synthesis protocols with accurate control of the magnetoelectric nanocomposites with satisfactory biocompatibility.[96] Moreover, further investigations into the therapeutic applications are needed to match the estimated in vivo conditions. In addition, multiferroic composite materials, which provide unique opportunities as sensitive sensors to measure ultra-weak magnetic fields,[97] are worth exploring in a large variety of biomedical field.

In summary, although multiferroic nanocatalytic materials provides an exciting opportunity for magnetically driven catalytic applications, the development of this new area remains at the initial stage with many critical and challenging issues. More studies on materials design and engineering, mechanism, and applications are highly expected. We hope this review will help researchers to gain an overview of this new field and inspire more innovative concepts to push forward the advancement in both fundamental science and practical implementation of magnetically driven catalysis using multiferroic nanomaterials.

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■ CONFLICT OF INTEREST

The authors declare no conflict of interest.

■ AUTHOR CONTRIBUTIONS

X. Zhang, Y. Zhang and F. Yang wrote original draft; J. Zhang, Y. Liu and S. Li supervised this work and helped with revisions. All authors had approved the final version.

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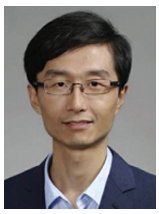
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