



Contact-electro-catalysis

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Charge transfer at the solid–liquid interface is essential in various systems, including catalysis, energy storage, and biological processes. Recent studies show that the contact between solid and liquid will lead to triboelectricity, electrons transferring, and catalyzing redox reactions, named contact-electro-catalysis (CEC). CEC uses materials such as hydrophobic polymers and inorganic compounds that were previously seen as inert, broadening catalyst selection. Additionally, powered by mechanical energy, CEC has a broad reaction domain and is capable of utilizing wasted energy, reducing reliance on fossil fuels and cutting carbon emissions, making it significant for advancing green chemistry. In this article, we will discuss the mechanism and potential applications of CEC.

Introduction

Charge transfer at the solid–liquid interface is a fundamental phenomenon observed across various systems, including heterogeneous catalysis, electrochemical energy storage and conversion, and biological processes involving cell membranes and tissue fluids.^{1,2} Therefore, the study of charge transfer at the solid–liquid interface holds significant importance for fundamental science as well as applications in energy and chemical industries.^{3,4} Recent studies have revealed that solid–liquid contact leads to spontaneous contact electrification (also known as triboelectricity), resulting in the transfer of electrons between the solid and liquid phases.^{5,6} This electron transfer not only catalyzes the generation of reactive free radicals and the reduction of metals, but also drives a variety of redox reactions, collectively referred to as contact-electro-catalysis (CEC).

CEC systems typically employ solid–liquid contact electrification, where the solid materials are typically hydrophobic organic polymers^{7–9} (e.g., FEP, PTFE) or strongly hydrophobic inorganic compounds (e.g., SiO₂, Al₂O₃). In the past, these materials were often considered chemically inert, but under the newly established CEC mechanism, they have emerged as viable catalysts for CEC reactions.^{10–12} Moreover, the energy source for CEC is mechanical energy.^{13,14} On one hand, the transmission of mechanical energy is less obstructed compared to electricity and light, thus allowing CEC to have a deeper and broader effective reaction domain; on the other hand, the abundance of wasted mechanical energy in the environment provides a sustainable energy source for CEC reactions. By harnessing mechanical energy, CEC offers the potential to reduce reliance on fossil fuels and electricity, thereby contributing to substantial reductions in carbon emissions within the chemical industry. Consequently, CEC not only broadens the scope of catalyst selection, but also demonstrates considerable scientific and practical significance, particularly in advancing sustainable chemical processes (**Figure 1**).

Mechanism of CEC

CEC is proposed based on the coupling of the physical and chemical process in the electron transfer during solid–liquid contact electrification (CE), representing a typical interdisciplinary research field.^{10,15} Taking the interaction between FEP

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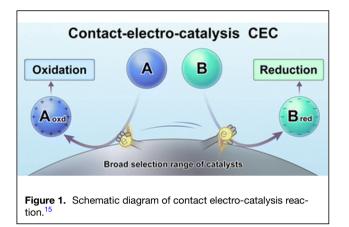
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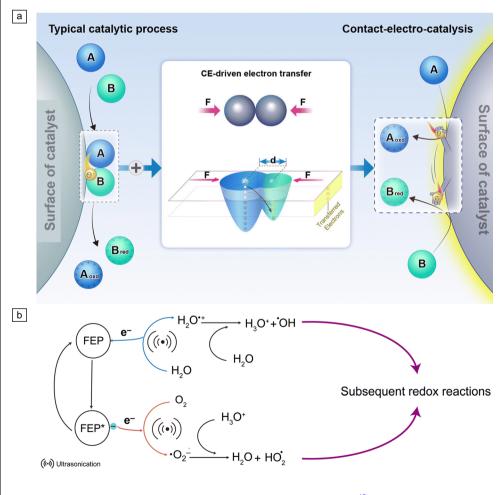
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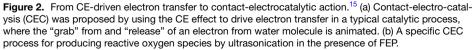
particles and an aqueous solution as an example, when water molecules come into contact with FEP particles, electrons are transferred from the water molecules to the FEP surface. Subsequently, dissolved oxygen in the water gains electrons from



the FEP surface, which is a physical process. From the chemical perspective, the water molecules lose electrons, converting water into radical cations, which then rapidly transfer protons to form hydroxide ions and hydroxyl radicals. As the electrons on the surface of the FEP particles are captured by O_2 molecules, superoxide radicals are formed, restoring the FEP particles to their original uncharged state. This cycle continues with the persistence of mechanical stimulation, generating a large number of reactive radicals in the solution, thereby promoting the occurrence of related chemical reactions, which is the basic principle of contact-electrocatalytic action (**Figure 2**).

It is evident that this process involves significant physicochemical intersection mechanisms, many of which warrant further in-depth investigation. For instance, Tang et al.¹⁶ employed a dynamic analysis of droplet sliding to investigate the phenomenon of charge transfer at the solid–liquid triboelectric interface. From a macroscopic statistical perspective, they reaffirmed that for hydrophobic solid materials, the primary charge carriers transferred during solid–liquid triboelectricity





are electrons. Furthermore, to better establish the correlation between electron transfer energy and the occurrence of chemical reactions, they proposed that during the solid-liquid CE process, electrons transfer from the frontier orbitals of liquid molecules to the frontier orbitals of solid material molecules,¹⁷ and then realize the relevant chemical reactions based on the potential of electrochemical reactions. Additionally, the process of exciting electron transfer involves the conversion of mechanical energy to electrical energy and then to chemical energy; hence, high-energy efficiency excitation methods are of significant importance. Beyond ultrasonic excitation, they have further proposed ball milling excitation¹¹ and anticipate expanding to stirring or vibration excitation. Moreover, although most materials possess triboelectric properties and can be used as catalysts in CEC, when it comes to solid–liquid reaction environments, materials are required to have both high triboelectricity and appropriate energy levels to facilitate subsequent chemical reactions. Tang and Wang's team proposed a polymer/metal Janus structure that combines the high triboelectricity of polymer materials, the mirror charge of metals, and the work function regulation of metals themselves, achieving a synergy between the water oxidation and oxygen reduction rates contained in CEC, thereby enhancing the reaction rate of CEC.¹⁸ More specific research is yet to be conducted in-depth.

Applications of CEC

CEC is realized by electron transfer during the mechano-driven contact electrification process, which differs from conventional catalytic principles in both electron and energy sources.^{15,19} Due to the significant electron transfer during the contact electrification process, and the fact that the charged surface after contact can locally introduce a strong electric field, these electrons combine with water or dissolved oxygen in water to generate active oxygen species such as hydroxyl radicals and superoxide radicals.²⁰ This promotes subsequent redox reactions and enables various applications. Moreover, the selection range for its catalysts is broad.^{15,21} As long as the material exhibits excellent contact electrification effects, such as commercially available polymers, it can be used for CEC. Furthermore, due to its environmentally friendly nature, mild reaction conditions, and other characteristics, it holds promise for exploring its applications in many catalytic reactions and even interdisciplinary fields, such as the degradation of organic pollutants, the synthesis of important chemicals, resource recovery, and even biomedicine. In this section, we will discuss the application of CEC in these representative directions.

Degradation of organic pollutants

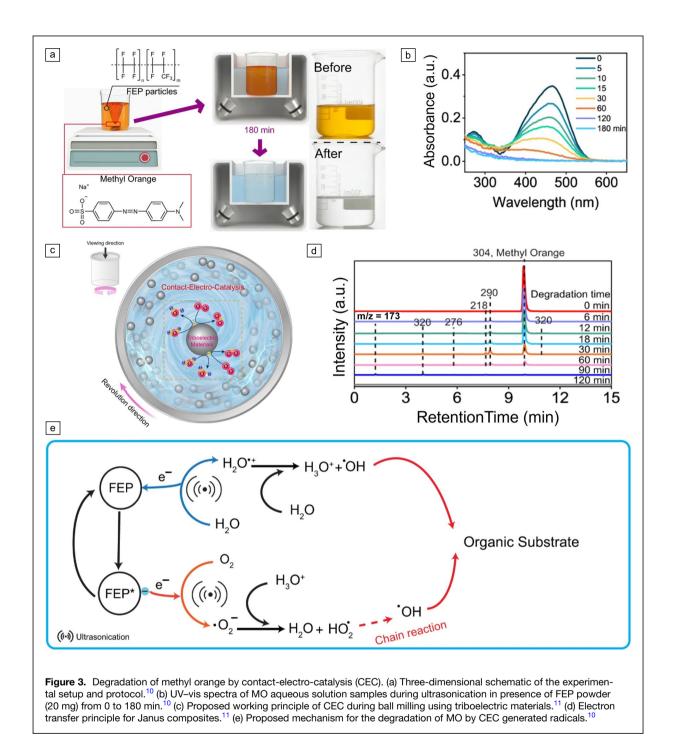
Organic pollutants commonly originate from industrial discharges, pharmaceuticals, pesticides, and other human activities. Their toxicity and potential for bioaccumulation pose significant threats to the environment and human health. Effective degradation of these pollutants helps prevent ecosystem disruption and reduces health risks, making it essential for safeguarding environmental health and protecting water and soil resources.^{22,23}

CEC is able to utilize the ambient mechanical energy to cause a contact electrification effect between materials to generate reactive oxygen species (ROS), enabling the efficient degradation of organic pollutants and other hazardous substances. The first example is the catalytic degradation of methyl orange (MO) as shown in **Figure 3**a. In this study, 20 mg of FEP powder was added to 50 mL of an aqueous MO solution (5 ppm) and stirred for 48 h to enhance the contact between FEP and water.¹⁰ Subsequently, the prepared suspension was subjected to ultrasonic treatment for 3 h, and the initial light-yellow solution became transparent as shown in Figure 3b. To investigate the discoloration process, the corresponding UV–vis spectra revealed that the characteristic absorption peak of MO gradually decreased with increasing ultrasonic treatment time, eventually

disappearing after 120 min. These results demonstrated that the MO solution was completely degraded within 3 h of ultrasonic treatment. To further understand the underlying mechanism, trapping experiments indicated that two reactive radicals, ·OH and $\cdot O_2^{-}$, were involved in the degradation process. Among these, hydroxyl radicals were identified as the primary limiting factor.¹⁰ Wang et al. further demonstrated that ball milling could directly promote frequent contact-separation cycles, with triboelectric materials catalyzing the generation of ROS to interact with organic pollutants in aqueous solutions. A ball milling setup was constructed (Figure 3c), where 50 mL of a 5 ppm MO solution was milled with 100 g of polymer balls for 120 min. The degradation rates of MO were 3.1%, 32.05%, and 99.18% for polypropylene (PP), PDMS, and PTFE, respectively. Then, liquid chromatography-mass spectrometry (LC-MS) was performed, as shown in Figure 3d. The characteristic absorbance intensity of MO in the UV-vis spectrum decreased with increasing milling time, approaching zero after 60 min. The evolution of MO's relative concentration in the presence of different scavengers revealed the contributions of ·OH and superoxide radicals $\cdot O_2^-$ to the degradation process. Additionally, the study investigated changes in MO's relative concentration under varying rotational speeds using PTFE. When the rotational speed was below 100 RPM, MO degradation was negligible. However, at 150 RPM, the relative concentration of MO dropped 19.5-fold, and further increases in rotational speed resulted in even higher degradation rates.¹¹

Synthesis of vital chemicals

The second example is the synthesis of vital chemicals. Hydrogen peroxide (H₂O₂) presents significant potential for environmental remediation and as an alternative to carbonbased energy sources.²⁴ As a green and mild oxidizing agent, hydrogen peroxide is a crucial chemical raw material with a wide range of applications in chemical synthesis, fuel energy, and the electronics industry.^{25,26} In recent studies, Zhao²⁷ has proposed the use of poly(tetrafluoroethylene) (PTFE) particles in contact electrolysis for the efficient generation of hydrogen peroxide (H_2O_2) from water as shown in Figure 4a. They achieved an H₂O₂ production rate of 313 μ mol L⁻¹ h⁻¹ under ambient temperature and pressure conditions, while Wang et al. enhanced the H_2O_2 production rate to 24.8 mmol $g_{at}^{-1} h^{-1}$ by utilizing hydroxyl radicals (·OH) generated from water oxidation. This study achieved high H2O2 generation rates. The CEC catalytic generation of hydrogen peroxide can be divided into two pathways: water oxidation reaction (WOR) and oxygen reduction reaction $(ORR)^{27,28}$ (Figure 4a). In the water oxidation reaction, PTFE particles come into contact with water, causing the water molecules to lose an electron to the PTFE surface, which generates a hydroxyl radical. The recombination of two hydroxyl radicals then produces a hydrogen peroxide molecule. In the oxygen reduction reaction, the solution undergoes ultrasonic treatment, generating numerous small bubbles that expand and then collapse. Oxygen molecules gain electrons from the charged PTFE surface, forming



superoxide radicals. The generated superoxide radicals are first protonated to form hydroperoxyl radicals (\cdot OOH), which then simultaneously gain a proton and an electron, transforming into hydrogen peroxide. At this point, the PTFE particles return to their original state. The \cdot OH as a highly oxidizing reactive oxygen species is considered an important oxidant for the oxidation of small gaseous molecules such as methane and nitrogen, in addition to its own combination into H₂O₂. Based on the CEC mechanism, Li et al. used air and methane as reactant gases and FEP as the catalyst to achieve the conversion of CH_4 to HCHO and CH_3OH under ambient conditions, with yields of 467.5 and 151.2 µmol gcat⁻¹, respectively.²⁹

Additionally, ammonia (NH₃) is one of the most important inorganic chemicals in industry. It is well known that nitrogen (N₂) makes up approximately 78.8% of the Earth's atmosphere, making the conversion of this abundant nitrogen source into ammonia a key focus of current research.³⁰ Zare introduced nitrogen gas into pure water containing poly(tetrafluoroethylene) (PTFE) particles at room temperature and synthesized ammonia (NH₃) via CEC principles with ultrasound as shown in

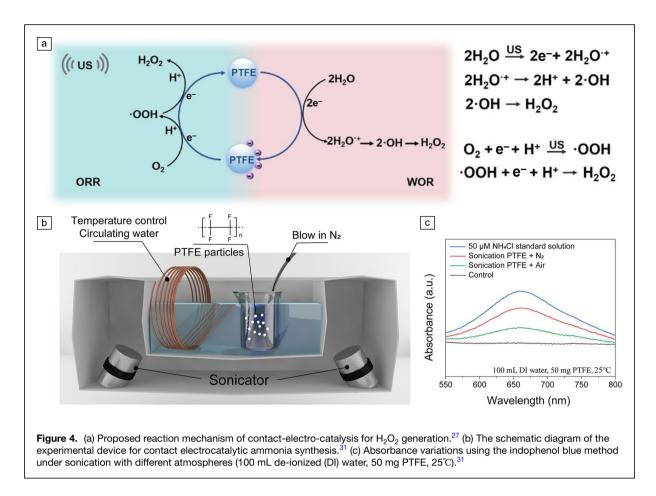


Figure 4b.³¹ Under optimal conditions, Li et al. synthesized ammonia (NH₃) by bubbling nitrogen (N₂) gas into bulk liquid water (200 mL) containing 50 mg poly(tetrafluoroethylene) (PTFE) particles (~5 μ m in diameter) suspended with the help of a surfactant (Tween 20,~0.05 vol%) at room temperature (25°C), and the ammonia production rate per gram of PTFE particles was approximately 420 μ mol L⁻¹ h⁻¹. As shown in Figure 4c, nitrogen gas injection under 2 h of ultrasonic treatment was more favorable for ammonia generation compared to air injection. Li et al. proposed a possible reaction mechanism for ammonia formation, suggesting that the ammonia synthesis reaction triggered by the contact electrolysis process can be divided into three steps.

First, water forms hydrogen ions and hydroxide ions at the interface. At the same time, nitrogen is adsorbed on the surface of PTFE, as shown in reactions (Equations 1 and 2):

$$H_2O \rightarrow H^+ + OH^-$$
 1

$$PTFE + N_2 \rightarrow PTFE-N_2.$$
 2

Second, promoted by the strong electric field caused by contact electrification at the interface, the hydroxide ion loses an electron to form a hydroxyl radical. Some electrons are trapped by hydrogen ions to form protons (hydrogen radicals), as shown in reactions (Equations 3 and 4):

$$OH^- \rightarrow OH^- + e^-$$
 3

$$\mathrm{H}^{+} + \mathrm{e}^{-} \to \mathrm{H}^{\cdot}. \qquad 4$$

Finally, H' primarily reacts with the adsorbed nitrogen (PTFE-N₂) in a series of steps to form NH_3 , which escapes from the PTFE to regenerate it, as shown in reaction (Equation 5). In addition, hydroxyl radicals recombine to hydrogen peroxide, as shown in reaction (Equation 6):

$$PTFE-N_2 + 6H^{\cdot} \rightarrow 2NH_3 + PTFE \qquad 5$$

$$OH^{\cdot} + OH^{\cdot} \rightarrow H_2O_2.$$
 6

Resources recovery

CEC is shown to be useful in resource recovery, including ithium-ion batteries (LIBs) and precious metals. The flowchart of LIBs recovery by CEC leaching is shown in **Figure 5**a.³² The process begins with pretreating the collected spent batteries to obtain lithium cobalt oxide (LCO) electrode powder. This is followed by CEC leaching, where citric acid serves as the leaching agent, and recyclable SiO₂ is added as a catalyst. The electrode powder is mixed with a solid-to-liquid ratio of 10 g L⁻¹, and ultrasonic stimulation is applied. The ultrasound generates cavitation bubbles, which collapse, causing frequent contact between SiO₂ and the aqueous phase. This results in the transfer of electrons from de-ionized water to the SiO₂ surface. Hydroxyl radicals (·OH) generated by the reaction

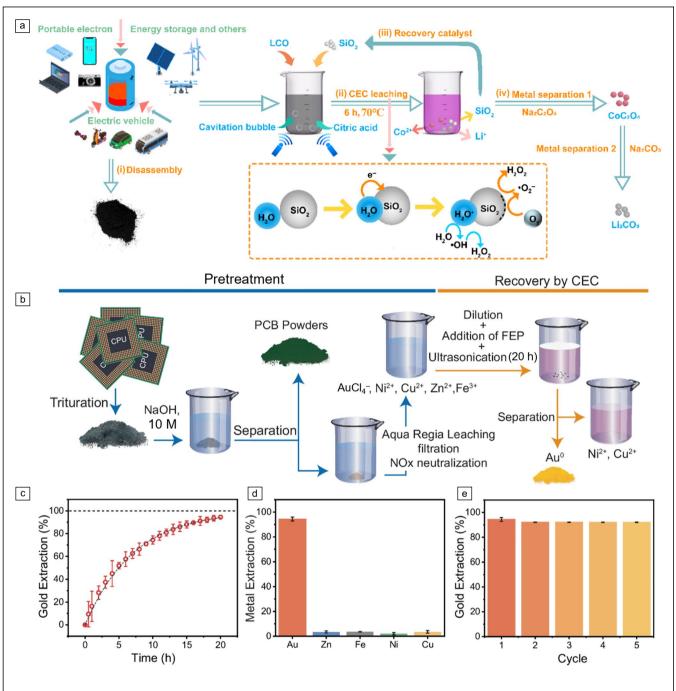


Figure 5. (a) Flowchart of lithium battery recovery by contact-electro-catalysis (CEC) leaching.³² Selective extraction of gold from the leachate of spent central processing units (CPUs) by CEC. (b) Schematic of the pretreatment of CPU (blue) and recovery of CEC for extracting gold from spent CPUs. Arrows are guide to eyes.³³ (c) Evolution of gold extraction over time from a leachate of CPU at a concentration of 10 ppm of gold.³³ The dotted lines are guides to the eyes. (d) Amount of $AuCl_4^-$, Zn^{2+} , Fe^{2+} , Ni^{2+} , and Cu^{2+} extracted after 20 h.³³ (e) Recycling of FEP particle for five cycles of 18 h. Error bars represent standard deviations for three reproduced experiments.³³

of water cations with water molecules break the Co–O bond, facilitating the leaching reaction. Electrons on the SiO₂ surface reduce Co^{3+} to Co^{2+} , enabling the leaching of metals. Oxygen captures electrons from the SiO₂ surface, forming superoxide anions, and the leaching of metals from LCO can be described by the reactions in equations. More importantly, hydroxyl radicals can recombine to form hydrogen peroxide, while superoxide radicals can also react with electrons to generate hydrogen peroxide, thereby accelerating the leaching of metals. This process results in a pink solution containing lithium and cobalt ions. Inductively coupled plasma-optical emission spectroscopy (ICP-OES) analysis indicates that after 6 h of ultrasound at 90°C, the leaching efficiencies for lithium and cobalt are 100% and 92.19%, respectively.

Su et al. conducted pioneering research on the feasibility of using CEC to extract precious metals.³³ They proposed that, following ultrasonic-assisted water-solid contact electrification (CE), the charged fluorinated ethylene propylene (FEP*) emits electrons into the solution under the excitation provided by ultrasonic vibrations. These electrons can then reduce metal ions present in the solution. Once electrons depart from the FEP, it returns to its ground state (FEP), and as long as ultrasonic conditions are maintained, the catalytic cycle continues. Using CEC for metal reduction, elements such as Ir, Au, Pd, Hg, Rh, Pt, and Ag can be extracted from aqueous solutions. CEC can recover gold from synthetic solutions with concentrations ranging from as low as 0.196 ppm to as high as 196 ppm, achieving extraction capacities between 0.756 and 722.5 mg g^{-1} within 3 h. Furthermore, selective recovery of gold from e-waste was demonstrated as shown in Figure 5b, which is the flowchart of the recovery process using CEC. After pretreating recovered CPUs to obtain a 10-ppm gold solution, adding 10 mg of FEP under aerobic conditions at 25°C for 20 h resulted in 94.4% of AuCl₄⁻ being reduced to elemental gold as depicted in Figure 5c, with minimal reduction of other metal ions (Zn, Fe, Ni, Cu) as illustrated in Figure 5d, indicating high selectivity of the CEC process. A similar approach applied to electroplating waste achieved a 91.5% gold extraction rate. This selectivity is likely due to a kinetic mechanism that prevents the redissolution of metals by neutralizing the solution, thus avoiding reversible reactions. Additionally, as shown in Figure 5e, after five cycles, the gold recovery efficiency did not significantly decrease.

Biomedical applications

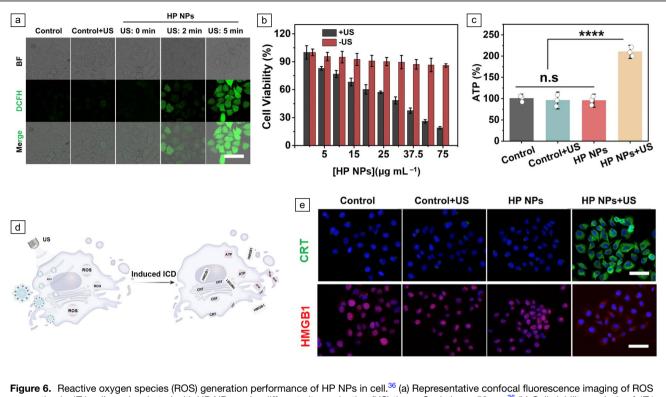
CEC harnesses interfacial electron transfer during contactseparation cycles of commercial dielectric materials with water, generating reactive oxygen species (ROS). However, the hydrophobicity of these materials limits reaction sites, and ROS often combine to form H₂O₂, reducing efficiency. To address this, Li et al. developed a PTFE-ZSM-5 catalyst (PZ) and an Fe^{III}-mediated Fenton-like system, achieving uniform catalyst dispersion and enhanced ROS generation.³⁴ This system enabled nearly 99% degradation of azo dyes within 10 min-a sixfold improvement over conventional CEC. Beyond environmental applications, CEC-generated ROS have shown promise in cancer therapies, such as sonodynamic therapy (SDT), chemodynamic therapy (CDT), and photodynamic therapy (PDT).³⁵ For example, Jiang et al. introduced a groundbreaking system utilizing a perfluorocarbon (PFC)-water "liquid-liquid" interface, where electron transfer processes during contact-electrification-induced interfacial redox reactions were shown to generate ROS.³⁶ PFC, known for its exceptional oxygen affinity, has been extensively employed to mitigate tumor hypoxia. In their study, human serum albumin (HSA) served as a stabilizing agent to fabricate PFC nanoemulsions (HP NPs).

Intracellular ROS levels were quantified using the ROSsensitive probe DCFH-DA, with fluorescence imaging by confocal laser scanning microscopy. As shown in Figure 6a, ROS levels increased significantly in HP NPs-treated cells upon ultrasonication, producing pronounced green fluorescence. Prolonged ultrasonication further amplified intracellular ROS levels, reaching 3.8 times that of the HP NPs-treated group without ultrasound (HP NPs+US 0 min group, US: ultrasound). ROS-mediated apoptosis was evaluated through MTT assays (Figure 6b), revealing that HP NPs, combined with ultrasound, significantly reduced the viability of 4T1 tumor cells due to ROS-induced cellular damage. Additionally, extracellular ATP release, a "find me" signal for attracting antigen-presenting cells (APCs), increased markedly in the HP NPs+US group (Figure 6c), indicating immunogenic cell death (ICD).

Contact-electrodynamic therapy (CEDT)-induced ICD was further supported by confocal imaging of high-mobility group box 1 (HMGB1) migration from the nucleus to the extracellular environment and calreticulin exposure on the cell membrane (Figure 6e). The HP NPs+US group exhibited the highest levels of CRT and HMGB1 expression, demonstrating strong green and red fluorescence signals, respectively. These "danger" signals are crucial for enhancing APC-mediated antigen presentation and activating systemic immune responses.

Conclusions

CEC is an emerging catalytic strategy that leverages contact electrification at interfaces to induce electron transfer, subsequently generating catalytically active species to drive redox reactions. This approach has demonstrated excellent performance across a wide range of applications, including pollutant degradation, CO2 capture, the synthesis of important chemicals (such as hydrogen peroxide and ammonia), resource recovery (e.g., recycling of lithium-ion batteries and precious metals), and even cancer therapy. CEC offers high reaction efficiency under mild conditions, which sets it apart from more demanding catalytic processes. More importantly, a particularly significant advantage of CEC lies in its broad catalyst selectivity. Unlike conventional photocatalysis, electrocatalysis, piezocatalysis, or tribocatalysis, which typically rely on semiconductors, metals, or carbon-based materials, CEC can utilize even chemically inert materials such as FEP and PTFE to achieve catalytic activity. This flexibility expands the range of potential catalysts significantly, making it easier to tailor catalytic systems to specific applications. Moreover, the scope of CEC extends beyond solid catalysts to liquid-liquid interfaces, such as oil-water boundaries, where liquid-liquid contact electrification can induce electron transfer to drive redox reactions, similar to solid-liquid systems. This capability opens up new possibilities for catalytic reactions that take place at liquid interfaces, where traditional catalytic methods could be less effective, thus offering a versatile and innovative pathway for advancing catalytic processes across various fields.



generation in 4T1 cells co-incubated with HP NPs under different ultrasonication (US) times. Scale bar = $50 \ \mu m.^{36}$ (b) Cell viability analysis of 4T1 incubated with different concentration of HP NPs with and without ultrasonication.³⁶ (c) Relative percentages of ATP in the extracellular environment of 4T1 cells under different treatments.³⁶ (d) Schematic representation of HP NPs-induced immunogenic cell death (ICD).³⁶ (e) Representative function of HP NPs in the extracellular environment of 4T1 cells under different treatments.³⁶ (d) Schematic representation of HP NPs-induced immunogenic cell death (ICD).³⁶ (e) Representative function of HP NPs in the extracellular environment of 4T1 cells under different treatments. Scale bar = $50 \ \mu m.$ CRT, calreticulin; HMGB1, high-mobility group box 1; ATP, adenosine triphosphate.³⁶

Currently, the greatest challenge limiting the development of CEC lies in its stringent requirements for reaction conditions. Effective CEC performance depends on strong contact electrification between the solid and liquid phases, which is typically optimized under neutral pH and low ionic concentration. This limitation results in lower efficiency when treating highly concentrated pollutants or operating in solutions with strong acidity or alkalinity. To address this issue, there is a need to explore more suitable catalysts that can meet the demands of specific catalytic reactions. Another potential approach is to incorporate other materials to enhance catalytic performance, thereby expanding the applicability of CEC. Additionally, improving charge density can further enhance catalytic efficiency, thereby reducing the constraints on reaction conditions. These strategies aim to enhance contact electrification capabilities, thereby improving the overall catalytic efficiency of CEC systems. Furthermore, a deeper exploration is needed to clarify the catalytic mechanism.

In summary, CEC offers exciting opportunities to advance mechanochemical catalysis. By modifying catalysts and gaining a deeper understanding of the underlying mechanisms, it is possible to discover novel catalysts and expand the scope of CEC applications. This positions CEC as a promising strategy for future research and industrial applications.

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Author contributions

W.T. and Z.L.W. were responsible for funding acquisition. W.T., F.R.F., and Z.L.W. provided supervision. W.T., F.R.F., and A.B. prepared the original draft. W.T., F.R.F., and Z.L.W. reviewed and edited the manuscript. All authors discussed the results and commented on the manuscript.

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Competing interests

All the authors declare no competing interests.

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