

Concave gas–liquid interface geometry as an acoustic field modulator for enhancing contact-electro-catalysis

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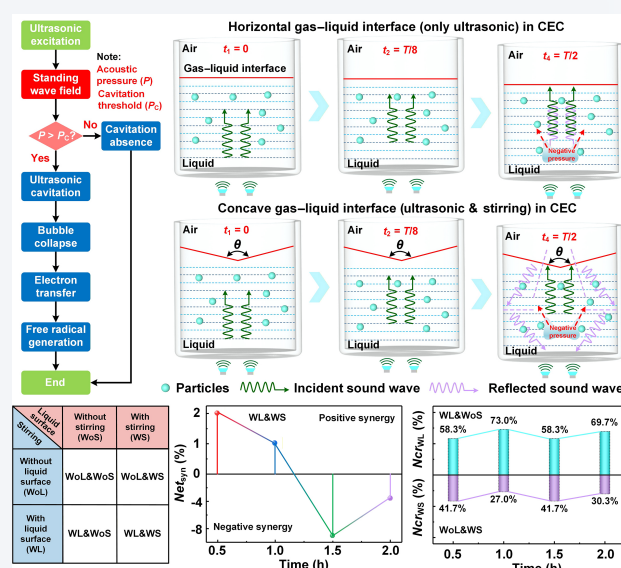
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ABSTRACT: Standing-wave-induced energy localization fundamentally limits the efficiency of ultrasound-driven contact-electro-catalysis (CEC), restricting effective coupling between mechanical excitation and catalytic reactions. Here, we report an acoustic field reconstruction mechanism in which the gas–liquid interface geometry functions as an active modulator of the acoustic field. A dynamically formed concave interface, generated through synchronized ultrasonic and stirring, redirects acoustic reflection pathways and disrupts standing-wave coherence. Acoustic simulations reveal that this curved interface enhances acoustic pressure amplitude and promotes a more homogeneous distribution of acoustic energy within the liquid phase. The reconstructed acoustic field significantly enhances catalytic performance across multiple reaction systems, including the degradation of representative organic molecules and the *in-situ* reduction of noble metal ions, following improved pseudo-first-order kinetics compared with conventional ultrasound-driven CEC. In addition to enhanced reactivity, reconstruction of the gas–liquid interface geometry reduces the overall energy consumption of the CEC system by approximately 32.99%. Importantly, this interfacial mechanism is independent of catalyst materials, highlighting the gas–liquid interface as a universal and reconfigurable functional element for regulating acoustic-electrochemical coupling. This work establishes an interfacial strategy for acoustic field manipulation, offering opportunities for the design of mechanically driven catalytic systems.

KEYWORDS: contact-electro-catalysis, standing wave structure, gas–liquid interface geometry, acoustic field reconstruction

1 Introduction

Mechanically driven catalysis has recently emerged as an attractive strategy for driving chemical reactions by converting mechanical energy into chemical reactivity. Among these approaches [1–3],



contact-electro-catalysis (CEC) has gained attention as an emerging mechanically driven catalytic mechanism [4–6]. This technology achieves electron transfer between materials through contact electrification, directly driving chemical reactions and demonstrating significant potential in the field of catalysis [7, 8]. Unlike traditional photocatalysis, electrocatalysis, or thermocatalysis, CEC avoids dependence on expensive precious metal catalysts, thus offering advantages such as low carbon footprint, environmental friendliness, mild conditions, and a wide range of catalyst material choices [9, 10]. The catalytic principle of

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CEC is based on contact electrification at the solid–liquid or liquid–liquid interface [11, 12]. Mechanical stimulation (such as ultrasound, ball milling, et al.) induces electron migration from the liquid phase to the solid material, thereby triggering redox processes, including water oxidation and oxygen reduction reactions, and forming highly reactive free radicals or promoting the reduction of metal ions [13–15]. Therefore, as long as the material has good contact electrification properties [16, 17], such as polymers like polytetrafluoroethylene (PTFE), fluorinated ethylene propylene (FEP), and poly(vinylidene fluoride) (PVDF), or inorganic materials like dielectric ceramics [18–20], it can participate in the reaction as a catalyst without having to meet the requirements of traditional catalysts for conductivity, semiconductor properties, or active metallic components.

Since the concept of CEC driven by ultrasonic cavitation was first proposed in 2022 [21], promoted its application in multiple fields. CEC can effectively generate reactive oxygen species, achieving efficient oxidation of recalcitrant organic pollutants, thus possessing significant value in environmental governance and wastewater treatment [22, 23]. Simultaneously, H_2O_2 can be synthesized at room temperature and pressure through simple material contact and ultrasonic cavitation, providing a more convenient technical route for green oxidation and wastewater treatment [24, 25]. Furthermore, CEC can also achieve the reduction and deposition of metal ions (such as Au and Pt), providing new solutions for resource recovery and precious metal extraction [26, 27]. With in-depth research, the academic community is also attempting to quantify the energy conversion efficiency of CEC and explore the influence of factors such as temperature, acoustic field parameters, and material properties on catalytic performance, laying a theoretical foundation for further optimization of its catalytic efficiency [28–30]. Meanwhile, the liquid–solid interface contact electrification and electrostatic double-layer modulation can promote charge transfer processes under mechanical perturbation [31, 32]. In the liquid–solid contact electrification, interface curvature affects charge transfer behavior, with convex surfaces being more conducive to charge transfer during droplet contact, while charge is more easily dissipated on concave surfaces [33–36]. Despite the rapid development of ultrasonic-driven CEC in recent years, it still faces a series of key challenges in practical applications. Traditional CEC relies heavily on ultrasound as the energy input mode, but the standing wave structure inevitably leads to excessive localization of acoustic energy in space, making it difficult for the reaction system to obtain a uniform and continuous energy supply [37, 38]. This acoustic field constraint determined by boundary conditions not only wastes a large amount of acoustic energy in ineffective regions but also significantly suppresses the spatiotemporal breadth of cavitation excitation, thus limiting the improvement of overall catalytic efficiency [39]. Although some studies have attempted to inject more acoustic energy into the system by increasing the frequency and power of the acoustic waves or by using multi-frequency coupling [40–42], the input energy is still difficult to be efficiently converted into usable chemical potential energy because the acoustic field topology itself has not been fundamentally changed, and the energy efficiency bottleneck of the system remains prominent. Therefore, maximizing the conversion efficiency of mechanical energy to chemical energy constitutes the core scientific problem and engineering challenge that urgently needs to be overcome in the current CEC field. In addition, CEC, as a universal catalytic technology that crosses

material systems, can be applied to a variety of catalytic materials, including polymers, ceramics, inorganic dielectric materials, and multiphase composite systems [43–45]. To achieve stable and efficient energy conversion in such diverse catalytic systems with significantly different physical properties, a universal acoustic field enhancement mechanism applicable across material systems is needed.

Herein, an acoustic field reconstructing mechanism based on actively controlling the geometry of the gas–liquid interface is proposed, fundamentally overcoming the bottleneck of low acoustic energy utilization efficiency caused by standing wave acoustic fields in traditional ultrasound-driven CEC alone. By designing the simultaneous ultrasonic and stirring (U&S) mode, the gas–liquid interface can be transformed from the static horizontal plane to dynamic concave interface. Acoustic simulations show that the change in the concave gas–liquid interface causes the acoustic wave reflection direction to be adjusted by the local normal, disrupting the spatial coherence of the standing wave, thereby redistributing the acoustic energy in the liquid phase and achieving a more uniform acoustic energy distribution. In experimental verification, this acoustic field reconstruction mechanism improves catalytic performance in both the catalytic degradation of various organic molecules as probes and the *in-situ* reduction of noble metals. Compared with CEC driven by ultrasound alone, the pseudo-first-order reaction kinetic constants are also improved. Simultaneously, the gas–liquid interface geometry reconstruction reduces the CEC overall energy consumption by approximately 32.99%. More importantly, this mechanism has material-independent universality, applicable to different solution systems and catalysts, and has good prospects for engineering applications. Therefore, the gas–liquid interface can serve as an active acoustic field modulator, providing a dimension of regulation for ultrasound-driven CEC, and offering an important theoretical basis and technical path for developing efficient, low-energy-consumption, and sustainable acoustic catalysis systems.

2 Experimental section

2.1 Preparation of organic molecular probe solutions

A methyl orange (MO, $\text{C}_{14}\text{H}_{14}\text{N}_3\text{NaO}_3\text{S}$, Macklin, 98%) solution was prepared by adding 5 mg of MO in 1000 mL of deionized water and stirring under magnetic stirring (700 r/min) for 2 h. The crystal violet (CV) and Rhodamine 6G (R6G) solutions were prepared as above.

2.2 Degradation of organic molecular probe solutions using the CEC system

First, 150 mL of the pre-prepared organic solution (R6G, MO, and CV) was transferred into a glass bottle. Then, 120 mg of catalyst powder (NR, PTFE, and SiO_2) was weighed into the solution and stirred magnetically for 48 h to ensure complete dissolution at 1000 r/min. Finally, the mixture was placed in an ultrasonic cleaner for ultrasonic (U) cavitation degradation for 120 min. During degradation, 3 mL of the mixture was extracted every 30 min and stored in a cylinder flask for subsequent analysis. The degradation steps for both only stirring (S) and simultaneous U&S were the same as described above, except that a stepper motor-driven PTFE stirring shaft was added, allowing ultrasonic and stirring (at speeds of 2.50, 3.75, and 5.00 r/s, respectively) to occur simultaneously, thus achieving mechanical coupling between ultrasonic and stirring.

2.3 Free radical quenching

Hydroxyl radicals ($\cdot\text{OH}$) quenching: The $\cdot\text{OH}$ radicals generated during ultrasonic cavitation were quenched by tert-butanol ($(\text{CH}_3)_3\text{COH}$). In short, 2 mL of $(\text{CH}_3)_3\text{COH}$ was added to a 150 mL solution containing PTFE and MO (after mixing). After stirring for 30 min, the mixture was placed in a simultaneous U&S device. During the degradation process, 3 mL samples were taken every 30 min for subsequent characterization and analysis.

Superoxide radical: The hydroquinone ($\text{C}_6\text{H}_4\text{O}_2$) was selected as the quencher for $\cdot\text{O}_2^-$. 2 mM $\text{C}_6\text{H}_4\text{O}_2$ was added to a 150 mL mixed solution of PTFE and MO. After stirring for 30 min, the mixture was placed in a simultaneous U&S device. During the degradation process, 3 mL samples were taken every 30 min for subsequent characterization.

2.4 Silver nitrate reduced by CEC

First, approximately 120 mg of PTFE particles were transferred to a glass bottle containing 150 mL of H_2O and magnetically stirred (1000 r/min) for 48 h. After stirring, 50 mg of AgNO_3 crystals were added to the mixture, and stirring was continued for 30 min until completely dissolved. Then, the glass bottle containing the mixture was placed in a simultaneous U&S device to reduce AgNO_3 , thus preparing Ag@PTFE composite material. After ultrasonic treatment, the mixture was allowed to stand for 24 h until precipitation occurred. After precipitation, the supernatant was aspirated with a pipette to obtain the Ag@PTFE composite material. Finally, 10 μL of the composite material and ciprofloxacin (CIP) (10^{-3} mol/L) solution were dropwise added to a PDMS flexible substrate using a pipette, and surface-enhanced Raman scattering (SERS) tests were performed under 785 nm laser irradiation.

2.5 Sample characterization

Scanning electron microscopy (SEM) was used to characterize the PTFE particles morphology. Transmission electron microscopy (TEM) was used to characterize the Ag@PTFE composites morphology and phase structure. Raman spectroscopy and Fourier transform infrared spectroscopy (FTIR) were used to characterize the PTFE and Ag@PTFE particles phase. An aliquot of the sample was characterized by ultraviolet-visible (UV-Vis) spectroscopy in the range of 300–800 nm.

3 Results and discussion

Figure 1(a) illustrates the core reaction pathway of electron transfer and free radical generation in the ultrasonic-driven contact-electrocatalysis (CEC) system. When ultrasound propagates in the liquid, if the acoustic pressure (P) amplitude reaches or exceeds the cavitation threshold (P_c), the cavitation causes the bubbles to collapse violently in a very short time, generating an instantaneous high-temperature and high-pressure environment [46–48]. This allows H_2O to contact and separate from inert polymer particles, accompanied by the transfer of interfacial electrons, then generating active free radicals such as $\cdot\text{OH}$, $\cdot\text{HO}_2$, and $\cdot\text{O}_2^-$. Figure 1(b) reveals the core physical logic of ultrasonic excitation in CEC. When $P > P_c$, the system enters the ultrasonic cavitation stage, followed by bubble collapse, electron transfer, and free radical generation. If $P < P_c$, the cavitation cannot form, and the related reactions cannot be initiated. This indicates that ultrasonic excitation is not an auxiliary step, but a decisive factor in improving the activity and

controllability of the CEC reaction. In conventional CEC systems, when ultrasound enters the liquid from the bottom of the reactor, the acoustic waves undergo specular reflection at the horizontal gas–liquid interface due to the acoustic impedance difference between the air and the liquid, resulting in “coherent superposition” of the incident and reflected waves [49]. As shown in Fig. 1(c), this boundary condition directly leads to the formation of a standing wave acoustic field, causing the acoustic pressure to exhibit a periodic peak-valley distribution in space. Since ultrasonic cavitation is only effectively excited near the acoustic pressure peak and almost impossible to occur in the acoustic pressure valley, this acoustic field segmentation effect caused by the geometry of the gas–liquid interface results in a huge energy waste, becoming a bottleneck for traditional U-driven CEC systems. To overcome the limitations of the standing wave-dominated acoustic field, the ultrasonic and stirring (U&S) mode is introduced to actively control the geometry of the gas–liquid interface. As shown in Fig. 1(d), under the action of rotational stirring, the originally static horizontal gas–liquid interface is reconstructed into a continuous dynamic concave interface. Compared with the horizontal interface, the concave interface causes the acoustic wave reflection to no longer occur in the original direction, but is controlled by the direction of the local normal of the interface, resulting in a high degree of spatial differentiation in the reflection behavior. Therefore, the incident and reflected waves no longer satisfy the coherent superposition condition for the formation of standing waves, thus weakening or even completely destroying the standing wave structure. From an acoustic physics perspective, this means that the gas–liquid interface transforms from a “plane mirror reflector” into a “curved surface scatterer”. This active modulation of the gas–liquid interface geometry triggers the topological reconstruction of the acoustic field. Acoustic energy is no longer confined to a few local regions but is redistributed to a wider angular domain, resulting in a nearly continuous, uniform, and high-intensity acoustic pressure environment within the liquid [49]. Figure 1(e) illustrates the limitations imposed by a traditional horizontal gas–liquid interface: strong standing waves, a single reflection direction, and a low-concentration acoustic field. In Fig. 1(f), the concave interface causes the reflection direction of the acoustic wave to be determined by the local interface normal, resulting in a dispersed trajectory of the reflected acoustic waves. The collinear superposition of the incident and reflected waves is broken, the standing wave topology is disintegrated, and the acoustic pressure field structure transitions from a “periodic segmented state” to a “continuous scattering state”. This transformation of interface geometry not only redistributes acoustic energy but also significantly enhances the spatial extent and duration of cavitation excitation, enabling cavitation to be activated simultaneously within a larger volume, rather than being limited to traditional standing wave antinodes. This mechanism fundamentally improves acoustic energy utilization efficiency, providing a physical basis for constructing an efficient, low-energy-consumption, and sustainable CEC reaction mode. From a more macroscopic scientific perspective, this discovery reveals that the geometry of the gas–liquid interface is an “invisible control parameter” of the acoustic field structure, rather than a passive boundary.

To verify the effect of gas–liquid interface shape on the modulation of standing wave acoustic field and acoustic pressure amplitude in CEC from an acoustic physics perspective, a three-dimensional (3D) acoustic simulation model is constructed to systematically examine the influence of different interface curvature

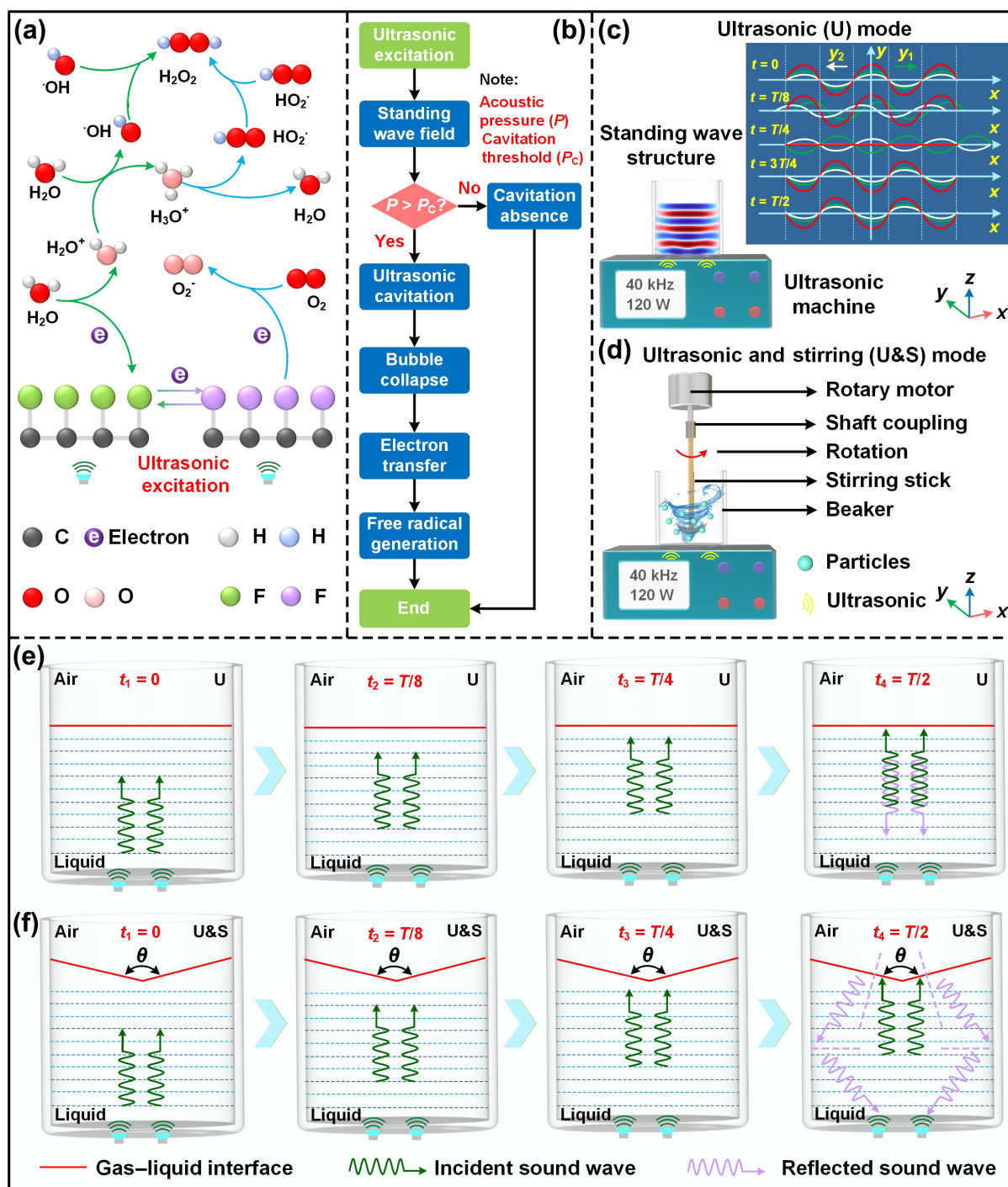


Figure 1 Ultrasonic field reconstruction mechanism based on gas-liquid interface regulation in CEC. (a) CEC working mechanism. (b) Logical framework of CEC working mechanism. (c) U mode. (d) U&S mode. (e) Acoustic wave reflection path under U mode. (f) Acoustic wave reflection path under U&S mode.

angles (θ) on the acoustic field structure. Figure S1 in the Electronic Supplementary Material (ESM) shows the 3D mesh model of gas-liquid interface shapes with different interface curvature angles. As shown in Fig. 2(a), by gradually adjusting the gas-liquid interface from $\theta = 180^\circ$ (horizontal interface) to $\theta = 140^\circ$, the dynamic transformation of the interface from horizontal to concave state under stirring can be simulated, thereby revealing the modulation mechanism of interface shape on the acoustic wave propagation path, reflection direction and final acoustic field distribution. As shown in Fig. 2(b), under the traditional horizontal interface

condition of $\theta = 180^\circ$, the reflection direction of the acoustic wave is very close to the incident direction, forming a typical and strong standing wave structure, with positive and negative acoustic pressure peaks of 0.97×10^7 and -1.01×10^7 Pa, respectively. In this case, the acoustic energy is confined to a local area, and the acoustic field exhibits high periodicity and spatial segmentation. Further, as shown in Fig. 2(c), when the interface angle slightly decreases to $\theta = 170^\circ$, the standing wave structure begins to weaken, and the positive and negative acoustic pressure amplitudes significantly increase to 1.26×10^7 and -1.22×10^7 Pa, respectively, corresponding to

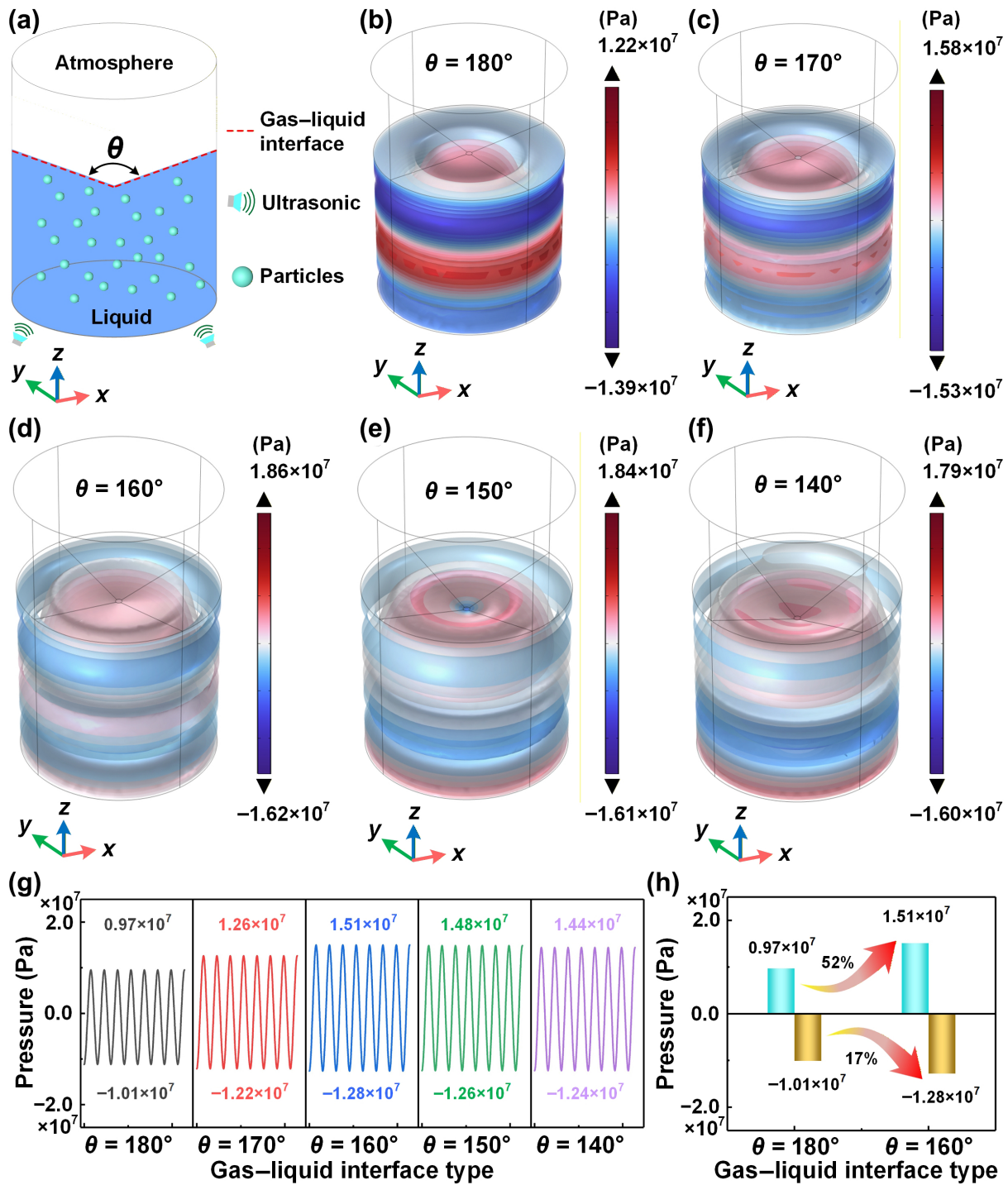


Figure 2 Effect of gas-liquid interface shape on acoustic field modulation. (a) Schematic diagram of the gas-liquid interface curvature angles (θ). (b) Acoustic field simulation at θ for 180° . (c) Acoustic field simulation at θ for 170° . (d) Acoustic field simulation at θ for 160° . (e) Acoustic field simulation at θ for 150° . (f) Acoustic field simulation at θ for 140° . (g) Acoustic wave amplitude comparison. (h) Acoustic wave amplitude comparison at θ for 0° and 160° .

increases of approximately 30% and 20%. This amplification phenomenon indicates that the acoustic pressure enhancement is not due to an increase in energy input, but rather to the reconstruction of the acoustic field boundary conditions. As the interface angle further develops towards concavity ($\theta = 160^\circ, 150^\circ, 140^\circ$; Figs. 2(d)–2(f)), the reflection behavior gradually changes from “quasi-specular reflection” to “scattering reflection” dominated by local normals. The reflected wave no longer returns

along the incident direction but is forced to redistribute to a wider angular domain, making it difficult for the incident and reflected waves to form a spatially coherent superposition, thus leading to the systematic destruction of the standing wave structure. This destruction does not damage the acoustic field energy but rather causes the acoustic field to enter a scattering-enhanced state. As shown in Fig. 2(g), when the interface angles are $\theta = 160^\circ, 150^\circ$, and 140° , the positive acoustic pressure amplitudes of the system

increase to 1.51×10^7 , 1.48×10^7 , and 1.44×10^7 Pa, respectively. Simultaneously, compared to the horizontal interface ($\theta = 180^\circ$), the corresponding negative acoustic pressure amplitude also first increases and then decreases as the interface angle decreases. As shown in Fig. S2 in the ESM, the experimental results at different θ generally show a generally consistent trend with the acoustic simulation results, that is, as the interface angle changes, the CEC degradation efficiency exhibits a pattern of first increasing and then decreasing. Further comparison of the acoustic pressure peak changes in Fig. 2(h) shows that when the interface bends to $\theta = 160^\circ$, the positive and negative acoustic pressure peaks are significantly increased by about 52% and 17%, respectively, compared to the horizontal interface. The above results clearly demonstrate that the concave gas-liquid interface, by introducing continuously varying local surface normals, causes a systematic shift in the acoustic wave reflection path, thereby disrupting standing wave coherence and driving a redistribution of acoustic flux across a larger angular domain [49]. This means that the acoustic field no longer exhibits the traditional standing wave structure composed of “nodes and antinodes”, but rather transforms into a multi-domain scattering acoustic field modulated by the interface geometry. Simultaneously, the gas-liquid interface is no longer a passive acoustic boundary but becomes an engineerable acoustic field modulator. As shown in Fig. S3 in the ESM, using a model with an interface angle of 160° as a representative example, three computational models with different mesh densities were constructed to compare the impact of mesh generation on the sound field simulation results. It can be seen that as the mesh is further refined, the peak values of positive and negative sound pressure levels and the overall sound pressure distribution do not change significantly, indicating that the simulation results are insensitive to mesh density and do not depend on a specific mesh generation.

To systematically evaluate the impact of gas-liquid interface shape modulation strategies on the actual activity of CEC, an U&S mode is constructed, as shown in Fig. 3(a). Using a series of organic molecules as reaction probes, the comprehensive comparisons are made between only U, only S, and the U&S conditions operating at different stirring intensities (2.50, 3.75, and 5.00 r/s). Experimental data in Fig. 3(b) show that only stirring system almost no degradation effect, while only ultrasonic system, although it can trigger a certain degree of cavitation, is still significantly constrained by the spatial limitations of the acoustic field. In stark contrast, the pseudo-first-order reaction kinetic constant of U&S system is significantly improved compared to the only ultrasonic system (0.91 h^{-1}) and only stirring system (0.00 h^{-1}). Notably, the U&S system exhibited the most significant catalytic acceleration at a stirring intensity of 3.75 r/s: its pseudo-first-order reaction kinetic constant increased from 0.91 h^{-1} under only ultrasonic conditions to 1.89 h^{-1} , representing an increase of more than 100%, as shown in Fig. 3(c). Figure S4 in the ESM illustrates the UV-Vis absorption spectra of MO solution degradation under U, S, and U&S-3.75 modes, respectively. It also further demonstrates that the concave shape of the gas-liquid interface brought about by ultrasonic stirring coupled mode is beneficial for improving the performance of CEC. Surface tension and viscosity are among the main physical factors affecting interfacial stability. Theoretically, if these parameters do not change significantly, the concave-convex interface remains macroscopically stable during solution degradation, as shown in Figs. S5 and S6 in the ESM. To clarify that the CEC kinetics

enhancement in the U&S system originates from the interface geometry reconstruction itself, rather than the additional effect of bubbles introduced during stirring. Meanwhile, to further elucidate the general mechanism of interface shape in acoustic field modulation, as shown in Fig. 3(d), four control systems are designed: (i) only U system, (ii) the planar Al sheet placed in the U solution (U&P) system, (iii) the stirring device introduced on top of the planar Al sheet (U&P&S) system, and (iv) the concave Al sheet placed in the U solution (U&C) system. The CEC activity is tested in these systems, as shown in Fig. 3(e). Compared to the U system, the U&P system exhibited higher catalytic degradation capacity. This is attributed to the superior reflective properties of the planar Al sheet compared to the gas-liquid interface, allowing more acoustic energy to be retained within the liquid rather than dissipating into the air. However, although stirring introduces additional bubbles in the U&P&S system, the catalytic degradation performance of CEC is essentially equivalent to that of U&P, indicating that bubble contribution is not the dominant factor in the enhanced kinetics. In contrast, the U&C system exhibits a significantly enhanced catalytic degradation capability. Because the concave Al sheet changes the local incident angle of the acoustic waves, it “decouples” the acoustic wave reflection from the original quasi-collinear directional pattern, thereby driving the redistribution of acoustic energy across a wider angular domain. Energy analysis further confirms this trend: Under U&C system, the energy consumption per unit time is reduced by approximately 32.99% compared to the U system (Fig. 3(e) inset), demonstrating a leap in energy efficiency resulting from acoustic field structure optimization. Figures S7 and S8 in the ESM illustrate the basis for energy consumption calculations under U and U&C modes. The specific energy consumption calculation values are shown in Table S1 in the ESM. This improvement of energy utilization efficiency is directly manifested in the reaction kinetics in Fig. 3(f): The pseudo-first-order rate constant of U&C reaches 1.68 h^{-1} , significantly higher than that of U (0.91 h^{-1}), U&P (1.09 h^{-1}), and U&P&S (1.13 h^{-1}). Figure S9 in the ESM shows the UV-Vis absorption spectra of MO solutions under different static interface modulation conditions. As shown in Fig. 3(g), the coupling relationship between interfacial concavity and stirring-induced fluid disturbance is further analyzed from the perspective of synergistic effects. By comparing four systems with and without liquid surface (WL/WoL), and with and without stirring (WS/WoS), the relative contributions of interfacial geometric control and stirring-induced fluid disturbance to CEC reaction kinetics can be clearly distinguished. Figure 3(h) shows that under WS&WL, the CEC brings a significant positive synergistic effect in the early and middle stages, but gradually transitions to a negative synergistic effect as the reaction progresses, suggesting that the contributions of the two types of factors change over time. This trend is further confirmed by the synergistic index analysis shown in Fig. 3(i): Under WL&WoS conditions, the concave interface effects net contribution rate (N_{crWL}) of the CEC remained high throughout the reaction process, which ranged from 58.3% to 73.0%. In contrast, the net contribution rate (N_{crWS}) of stirring-related effects introduced by stirring is only 27.0%–41.7%. The relative contributions of each factor to CEC performance and their specific values are shown in Tables S2–S4 in the ESM, including the net contributions of stirring-related effects and concave interface effects. This result indicates that although the mass transfer enhancement caused by stirring can promote catalytic degradation to some extent, its contribution is

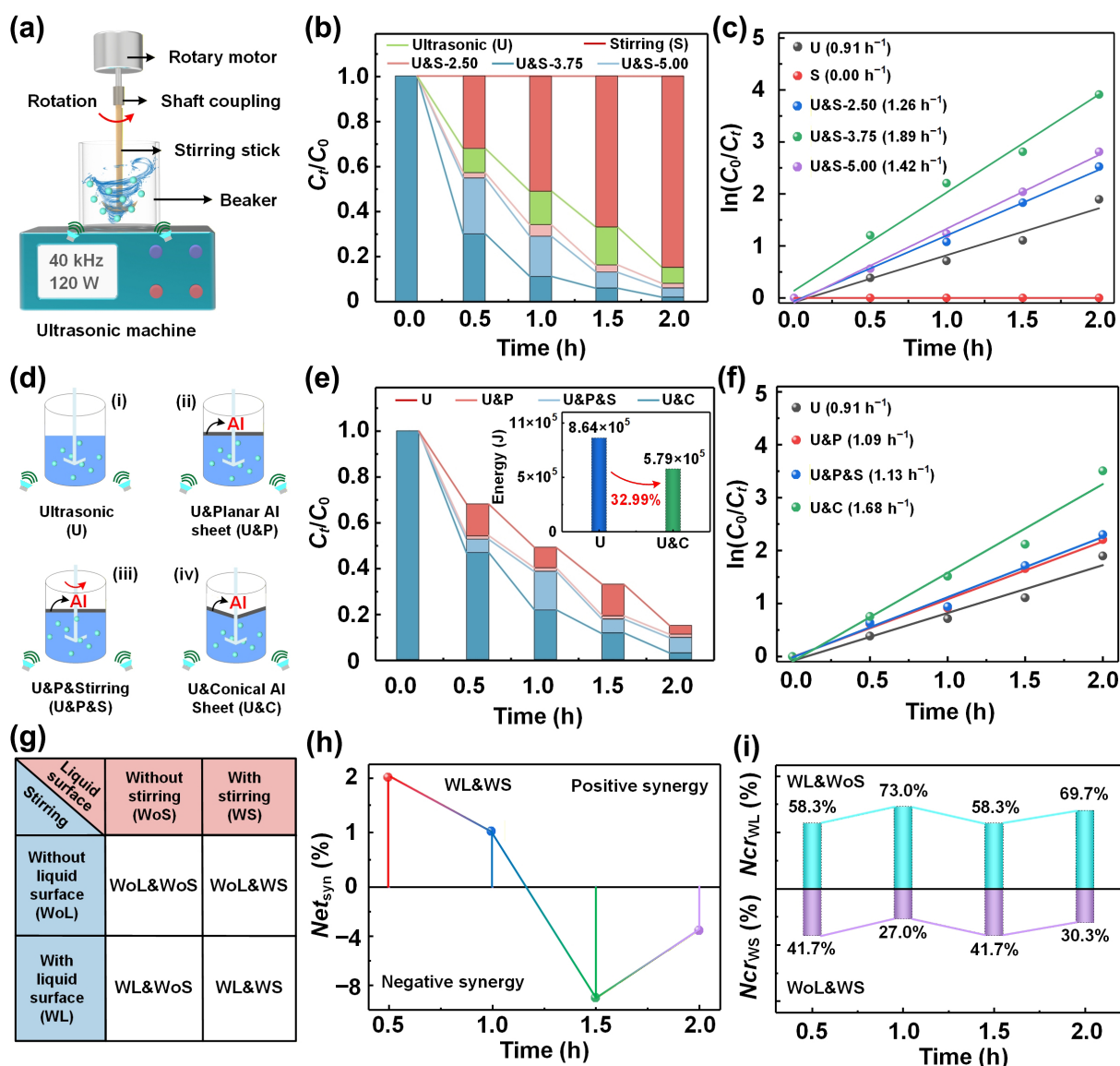


Figure 3 Experimental verification of the CEC performance enhanced by concave gas-liquid interface. (a) Device schematic diagram of U&S mode. (b) CEC performance under excitation modes of different mechanical energy. (c) Degradation kinetic fitting curves under excitation modes of different mechanical energy. (d) Device schematic diagram of CEC performance regulated by different static interface regulation conditions. (e) Effect of different static interfaces regulation conditions on CEC performance (insert: energy consumption comparison during the CEC process). (f) Degradation kinetic fitting curves under different static interfaces regulation conditions. (g) Collocation table of concave gas-liquid interface and stirring in CEC. (h) Synergy contribution of concave gas-liquid interface and stirring in CEC. (i) Individual contribution rates of concave gas-liquid interface and stirring in CEC.

significantly lower than the interfacial curvature effect produced by the concave interface. Furthermore, the impact of aluminum sheet material on the CEC catalytic performance in the system has been analyzed and discussed in detail in the supporting information and Tables S5–S7 in the ESM. Figure S10 and Table S8 in the ESM discuss the contact electrification phenomenon between the Al sheet and the cavitation bubbles and the possible influence of trace Al ions on the CEC performance, respectively, providing experimental basis for mechanism analysis. Figures S11 and S12 in the ESM also analyze and discuss the effects of convex interface conditions and the addition of an ultrasonic probe above the liquid surface on the catalytic performance of CEC.

To verify that the reconstruction of the acoustic field topology by the concave gas-liquid interface is a material-independent reaction kinetics enhancement mechanism, R6G, CV, and MO, which form

different solution systems, are selected as organic molecular probes. Simultaneously, using PTFE particles as an inert catalyst, the degradation effects of different solutions under U and U&S-3.75 are compared as shown in Figs. 4(a)–4(c), respectively. Figures S13–S15 in the ESM show the UV–Vis absorption spectra of R6G, CV, and MO solutions with PTFE as catalyst under different mechanical energy excitation modes. In all solution systems, the degradation kinetic constants (k) under U&S-3.75 is significantly higher than that under U, and compared with U, the degradation kinetic constants increased from 2.21 to 2.57 h⁻¹ (R6G), 1.49 to 1.91 h⁻¹ (CV), and 0.90 to 1.89 h⁻¹ (MO), respectively, as shown in Fig. 4(d). These kinetic constants increased by 16.29%, 28.19%, and 110%, respectively. Using MO as a molecular probe, the stability of PTFE as the catalyst under U&S-3.75 is investigated. Figure S16 in the ESM shows the C_0/C_t - t curves of MO solution degradation

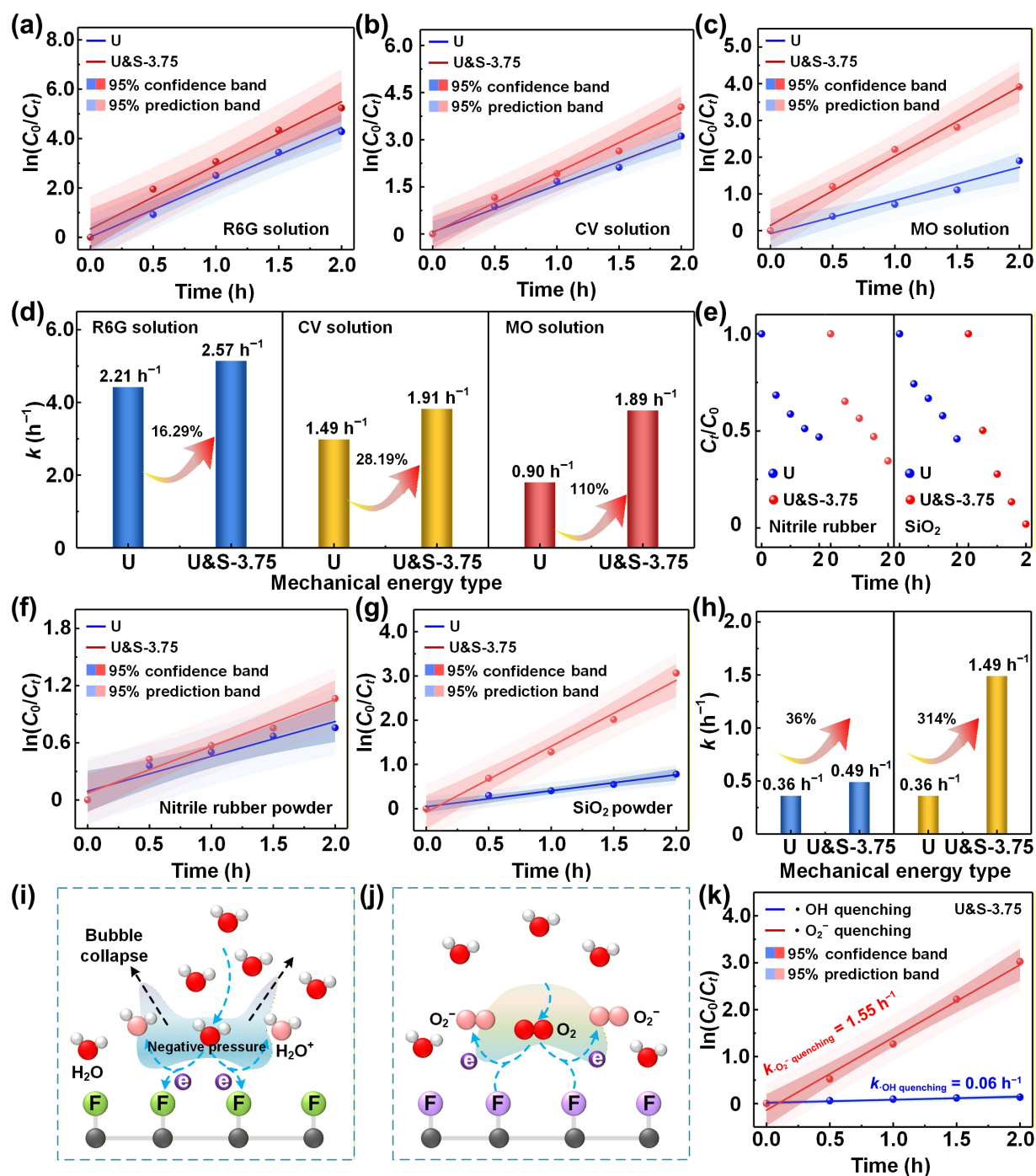


Figure 4 Material independence verification of CEC performance enhanced by concave gas-liquid interface. (a) Degradation kinetic fitting curves of R6G solution under excitation modes of different mechanical energy. (b) Degradation kinetic fitting curves of CV solution under excitation modes of different mechanical energy. (c) Degradation kinetic fitting curves of MO solution under excitation modes of different mechanical energy. (d) Degradation kinetic constant comparisons of R6G, CV, and MO solutions under excitation modes of different mechanical energy. (e) Degradation performance of nitrile rubber and SiO₂ powders as inert catalyst under excitation modes of different mechanical energy. (f) Degradation kinetic fitting curves of nitrile rubber powder as an inert catalyst. (g) Degradation kinetic fitting curves of SiO₂ powder as an inert catalyst. (h) Degradation kinetic constant comparisons of nitrile rubber and SiO₂ powders as inert catalyst under excitation modes of different mechanical energy. (i) Schematic illustration of CEC reaction for bubble collapse. (j) Schematic illustration of CEC reaction for bubble growth. (k) Comparison of reaction rates after adding different types of free radical quenchers.

under U&S-3.75, indicating that PTFE exhibits excellent catalytic stability. Figure S17 in the ESM shows the SEM characterization results of PTFE particles in the initial state, U, U&S, and after four cycles. The macroscopic morphology of PTFE particles in different states is shown in Fig. S18 in the ESM. Furthermore, Raman and

Fourier transform infrared spectra characterization results show that the phase structure of PTFE does not change significantly under different treatment conditions, as shown in Figs. S19 and S20 in the ESM, respectively, further confirming the good stability of PTFE as a catalyst. Additionally, to investigate the effects of

different solid particles on solution degradation, the nitrile rubber and SiO₂ particles are used to replace PTFE particles in MO solution, respectively. Figures S21 and S22 in the ESM show the UV–Vis absorption spectra of MO solutions using NR and SiO₂ as catalysts, respectively. The degradation effects of MO solution under U and U&S-3.75 are compared. Experimental results showed that under U&S-3.75 conditions, the concentration decay curves of MO solution (Fig. 4(e)) and its corresponding $\ln(C_0/C_t)-t$ fitting curves (Figs. 4(e) and 4(f)) both exhibited larger slopes and higher degradation kinetic constants. The nitrile rubber and SiO₂ particles have a significant impact on the solution degradation effect under U&S-3.75. As shown in Fig. 4(h), under ultrasonic stirring coupled conditions, the nitrile rubber particles increased the degradation kinetic constant from 0.36 to 0.49 h⁻¹ (an increase of 36%), while SiO₂ particles increased the degradation kinetic constant from 0.36 to 1.49 h⁻¹ (an increase of 314%). Compared with U, the degradation efficiency under U&S-3.75 conditions are improved, which verifies the significant effect of U&S-3.75 on enhancing the solution degradation rate in the CEC system. Therefore, the particle systems with different chemical compositions and interface properties all showed significant enhancement, indicating that the acoustic field-interface coupling mechanism does not depend on specific material properties, but is a general strategy for reshaping interface dynamics. It is worth emphasizing that this process is not only the passive generation of free radicals in traditional CEC system, but also an acoustic field-driven interface electronic engineering process, which makes CEC leap from “passive free radical chemistry” to “acoustic programmable interface electronic dynamics”. In the CEC system, cavitation bubbles undergo periodic growth and violent collapse. When the bubbles burst, water comes into contact with PTFE particles and becomes charged, forming an instantaneous potential difference, which drives electrons to transfer from the aqueous phase to the PTFE particles. PTFE gains electrons, while water molecules lose electrons to generate highly unstable ·H₂O⁺ free radical cations. This intermediate rapidly reacts with surrounding water molecules to produce hydroxyl radicals (·OH), as shown in Fig. 4(i). At the same time, in Fig. 4(j), the electrons captured on the PTFE surface can further react with dissolved oxygen to generate superoxide radicals (·O₂⁻). These highly reactive free radicals exhibit strong oxidizing capabilities and can effectively degrade various organic molecules in water, such as R6G, CV, and MO. Figure S23 in the ESM shows the UV–Vis absorption spectra of MO solutions after quenching by hydroxyl radicals and superoxide radicals. Figure 4(k) shows the degradation kinetic constants of MO solution under U&S-3.75 with the addition of ·O₂⁻ and ·OH radical quenchers (hydroquinone and tert-butanol), respectively. Experiments show that the apparent degradation rate constants of MO are: $k_{O_2^- \text{ quenching}} = 1.55 \text{ h}^{-1}$, $k_{OH \text{ quenching}} = 0.06 \text{ h}^{-1}$. The significant inhibitory effect of tert-butanol on the degradation process indicates that ·OH is the key reactive species dominating the degradation reaction; in contrast, although ·O₂⁻ participates in the reaction, its role is relatively minor. These results provide important guidance for a deeper understanding of the CEC reaction mechanism and the optimization of catalytic material design.

To further verify the universality and engineering value of the concave gas–liquid interface-enhanced acoustic field mechanism in the construction of noble metal materials, PTFE particles are used as catalysts to achieve reagent-free *in-situ* reduction of Ag⁺ through CEC system. Specifically, Fig. 5(a) shows the experimental setup for

in-situ deposition of silver ions on the PTFE surface under U&S-3.75 mode. This setup enhances the acoustic field through the concave gas–liquid interface, promoting ultrasonic cavitation and providing a higher-frequency and higher-energy microenvironment for metal nucleation. Figure 5(b) further reveals the mechanism of *in-situ* reduction of Ag⁺ particles during the CEC process. The ultrasonically driven cavitation microjets and collapse impacts induce frequent transient contact and charging between PTFE and H₂O molecules, thereby generating a considerable interfacial potential difference. This potential difference not only drives the rapid migration of electrons from the aqueous phase to the PTFE surface, but also activates water and dissolved oxygen to generate reactive free radicals such as ·OH and ·O₂⁻, enabling Ag⁺ to achieve *in-situ* reduction without an external power source or chemical reducing agent. This acoustic-triboelectric and free radical chemistry coupling pathway constitutes the core physical basis for the *in-situ* construction of noble metals. Figure S24 in the ESM shows photographs of Ag⁺ reduced solutions under different mechanical energy excitation modes. The results show that under only ultrasonic (U) mode, the Ag@PTFE samples are brown, while under the U&S-3.75 mode, the Ag@PTFE samples are black. This color difference indicates that there are significant differences in the degree of Ag⁺ reduction and the phase composition of the products under different mechanical energy excitation modes, and the U&S-3.75 mode being more conducive to the deep reduction of Ag⁺. Furthermore, the samples obtained after separating the reaction solutions and drying are shown in Fig. S25 in the ESM. At the material structure level, Fig. 5(c) shows the TEM of Ag@PTFE under U mode. It can be observed that the nucleation of Ag particles is relatively sparse and the particle size distribution is not uniform, indicating that although a single acoustic field can trigger the CEC process, its local energy supply is still limited. However, after treatment in U&S-3.75 mode, a large number of Ag-based particles with relatively narrow size distribution and uniform morphology are uniformly attached to the PTFE surface, as shown in Fig. 5(d)(i). Statistical results show that its average particle size is approximately 22.5 nm (Fig. 5(d)(ii)). The interplanar spacings of 0.227 and 0.208 nm in the TEM correspond to the characteristic crystal planes of Ag and Ag₂O, respectively (Fig. 5(d)(iii)), which are highly consistent with the PDF #41-1402 (Ag) and PDF #41-1104 (Ag₂O) diffraction peaks appearing in the XRD spectrum in Fig. 5(e). After drop-coating and drying the solution containing Ag@PTFE, a coffee ring structure Ag@PTFE SERS substrate can be obtained. As shown in Fig. 5(f), using typical ciprofloxacin (CIP) as the target molecule, the SERS spectrum of the Ag@PTFE substrate under U&S-3.75 mode showed significant enhancement throughout the fingerprint region, with a substantial increase in peak intensity compared to unloaded or inefficiently loaded Ag substrates. Compared to U mode, the enhancement factor of the Ag@PTFE SERS substrate under U&S-3.75 mode increased from 0.633×10^2 to 1.253×10^3 , corresponding to a relative increase of approximately 88.9%, as shown in Fig. 5(g), indicating that the concave interface effectively amplifies the electron utilization efficiency and metal nucleation density during the CEC process. More importantly, the SERS mapping shows that the signal spatial distribution at the characteristic peaks of 1408 and 1644 cm⁻¹ is extremely uniform, with RSDs of only 9.2% and 12.4%, respectively (Figs. 5(h) and 5(i)). This indicates that the acoustic field reconstruction not only improves the substrate enhancement factor (EF), but also statistically improves the uniformity of hotspot

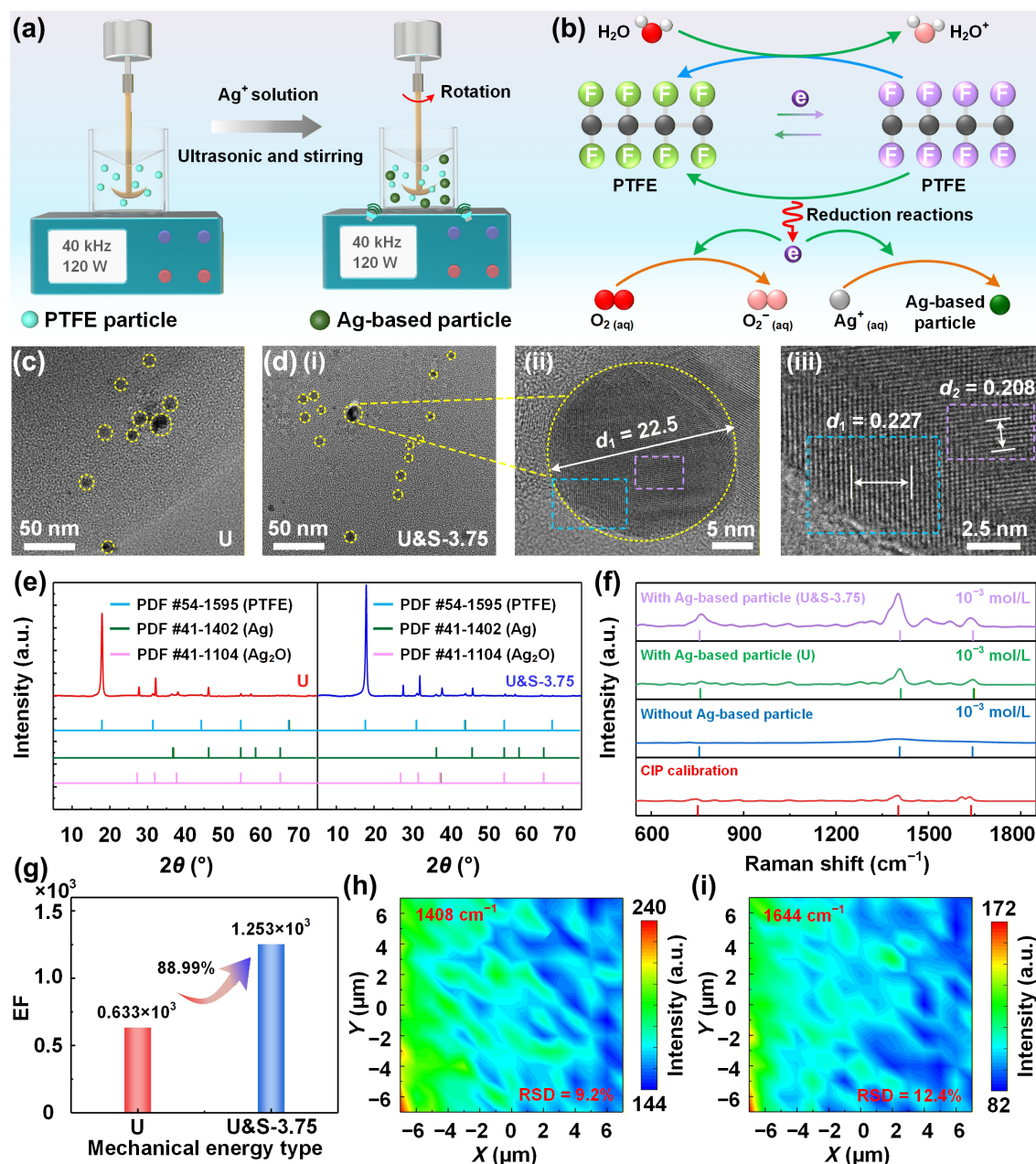


Figure 5 CEC-reduced Ag@PTFE based on concave gas-liquid interface regulation for SERS. (a) U&S mode for Ag@PTFE reduction. (b) Reduction mechanism of Ag@PTFE. (c) TEM image of Ag@PTFE reduced by U mode. (d) TEM image of Ag@PTFE reduced by U&S mode. (e) XRD curves of Ag@PTFE. (f) SERS curves for CIP detection. (g) SERS enhancement factor for CIP detection. (h) Raman mapping of Ag@PTFE substrate at 1408 cm⁻¹. (i) Raman mapping of Ag@PTFE substrate at 1644 cm⁻¹.

distribution, laying the foundation for highly reproducible spectroscopic detection. Therefore, the reconstruction of the gas-liquid interface geometry, and thus the control of the acoustic pressure energy distribution, enable CEC to exhibit high consistency and superiority in both nanoscale material construction and macroscale SERS applications, laying an important foundation for future expansion into Au, Pt and other multimetallic systems and their optical and catalytic functional materials.

4 Conclusions

This work proposes an acoustic field reconstruction mechanism based on actively controlling the gas-liquid interface geometry, and

achieves active modulation of the acoustic field propagation mode by designing an ultrasonic and stirring mode, thereby enhancing CEC efficiency. Compared with the traditional standing wave-dominated acoustic field, the dynamic concave interface can disrupt the coherent superposition of incident and reflected waves, causing the acoustic field to transition from a highly segmented periodic structure to a continuous scattering state, and thus improving the uniformity of acoustic energy distribution. This acoustic field reconstruction mechanism not only overcomes the inherent limitations of energy localization and low utilization caused by standing waves, but also achieves a more efficient conversion of mechanical energy into chemical energy. More importantly, the acoustic field modulation mechanism based on the gas-liquid

interface geometry has universal applicability across material systems, and can adapt to the complex requirements of different media acoustic impedances and various catalytic materials (including polymers, ceramics, inorganic dielectrics, and composite systems). Therefore, the gas–liquid interface can serve as an active acoustic field modulator, providing a new dimension of regulation for ultrasonically driven CEC, and opening up a development direction for acoustically driven catalysis technology towards programmability, tunability, and industrial application.

Electronic Supplementary Material: Supplementary material (Figs. S1–S25 and Tables S1–S8) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908886>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

J. F. M.: Data curation, validation, experimental design. M. Z. M.: Validation, experimental design. C. L. S.: Writing manuscript. J. M.: Validation. Y. Q. Q.: Data curation. Y. X. D.: Experimental design. W. X. L.: Data curation. Y. H. W.: Data curation. H. J. X.: Experimental design. S. Z.: Project administration, funding acquisition, writing manuscript, experimental design. All the authors have approved the final manuscript.

Use of AI statement

None.

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