

# Supporting Information

## Photochemical-Co-Mechanical Polishing for Silicon Wafer Using a Janus CeO<sub>2</sub> Slurry

Yuhang Qi<sup>1\*</sup>, Jinghui Lai<sup>1</sup>, Zihan Luo<sup>2</sup>, Shengying Yue<sup>2\*</sup>, Yuan Li<sup>3</sup>, Shiguang Guo<sup>1</sup>,  
Ziyang Li<sup>1</sup>, Guofeng Pan<sup>1</sup>, Xueli Yang<sup>1</sup>, Chong Luo<sup>1</sup>, Shuxin Ouyang<sup>3\*</sup>

<sup>1</sup>Tianjin Key Laboratory of Electronic Materials and Devices, School of Electronics  
and Information Engineering, Hebei University of Technology, Tianjin 300401, China

<sup>2</sup>State Key Laboratory for Strength and Vibration of Mechanical Structures, School of  
Aerospace, Xi'an Jiaotong University, Xi'an 710049, China

<sup>3</sup>Engineering Research Center of Photoenergy Utilization for Pollution Control and  
Carbon Reduction, College of Chemistry, Central China Normal University, Wuhan,  
430079 China

## Supplementary Methods

### Materials

All solutions used in this study were prepared with deionized water, and the abrasive was cerium oxide (99.95% purity, supplied by Shanghai Aladdin Biochemical Technology Co., Ltd.). All chemical reagents were of analytical grade. Unless otherwise specified, all solutions in this paper have a pH of 10. Chemical agents utilized encompass: hydrogen peroxide ( $\text{H}_2\text{O}_2$ ; with a purity of 30%, supplied by Tianjin Fengchuan Chemical Reagent Technology Co., Ltd.), PVP 99% pure, supplied by Shanghai Aladdin Biochemical Technology Co., Ltd.), silica sol (40% purity, supplied by Jinwei Group Co., Ltd.). Titanium Oxide ( $\text{TiO}_2$ ; 99.9% pure, supplied by Shanghai Aladdin Biochemical Technology Co., Ltd.), aluminium oxide ( $\text{Al}_2\text{O}_3$ ; 99% pure, supplied by Shanghai Dibo Chemicals Technology Co., Ltd.), ferric oxide ( $\text{Fe}_2\text{O}_3$ ; 99.5% pure, supplied by Shanghai Dibo Chemicals Technology Co., Ltd.), zinc oxide ( $\text{ZnO}$ ; 99.9% pure, supplied by Meryer (Shanghai) Biochemical Technology Co., Ltd.), strontium titanate ( $\text{SrTiO}_3$ ; 99% pure, supplied by Bide Pharmatech Co., Ltd.), tungsten trioxide ( $\text{WO}_3$ ; 99.8% pure, supplied by Anhui Zesheng Technology Co., Ltd.), stannic oxide ( $\text{SnO}_2$ ; 99.5% pure, supplied by Shanghai Macklin Biochemical Technology Co., Ltd.), cadmium oxide ( $\text{CdO}$ ; 99.99% pure, supplied by Beijing innoChem Science & Technology Co., Ltd.), zirconium dioxide ( $\text{ZrO}_2$ ; 99.99% pure, supplied by Mreda Technology Co., Ltd.), benzoquinone (BQ,  $\text{C}_6\text{H}_4\text{O}_2$ ; 99% pure, supplied by Mreda Technology Co., Ltd.), methanol ( $\text{MeOH}$ ,  $\text{CH}_4\text{O}$ ; 99% pure, supplied by Shanghai Macklin Biochemical Technology Co., Ltd.), perfluorosulfonic acid-PTFE copolymer (nafion; 5% pure, supplied by E.I.Du Pont Company), potassium hydroxide

(KOH; 99.9% pure, supplied by Shanghai Aladdin Biochemical Technology Co., Ltd.).

## **Electrochemical Experiment**

All electrochemical tests were performed using the CHI660E electrochemical workstation from Shanghai Chenhua Instruments Co., Ltd.

The corrosion properties of Si in designated solutions were investigated. Data for open circuit potential (OCP), dynamic potential polarization (Tafel plots), and impedance spectrum (EIS) were collected using a three-electrode system, with Si as the working electrode, standard calomel electrode (SCE) as the reference, and platinum as the counter electrode. Before each test, the Si electrode was sealed using insulating tape, revealing a 1 cm<sup>2</sup> active area. Before each electrochemical measurement, the samples were immersed in 5 wt% potassium hydroxide solution for 5 min to remove the natural oxide layer. This was followed by rinsing with deionized water and drying with compressed air. The OCP was stabilized for around 600 seconds to ensure the acquisition of trustworthy and consistent potential readings. Potentiodynamic polarization was measured within a voltage range of OCP  $\pm$  0.3 V at a scan rate of 5 mV/s. EIS was performed at the OCP, spanning a frequency range of 0.01 Hz to 1000 kHz, with a 5 mV AC signal applied. EIS data were analyzed and modeled using Zview software.

A uniform ink was prepared by dispersing 10 mg of the catalyst into 420  $\mu$ L of a solution containing 200  $\mu$ L of isopropanol, 200  $\mu$ L of deionized water, and 20  $\mu$ L of Nafion solution, followed by 30 minutes of ultrasonication. The resulting ink was evenly applied onto FTO glass,

with insulating tape to ensure exposure of a 1 cm<sup>2</sup> active area. Next, the prepared electrode was annealed at 200°C for 2 hours.

The transient photocurrent response measurements were conducted using a standard three-electrode system. Pt and Ag/AgCl electrodes served as the counter and reference electrodes, respectively, and the as-prepared electrode was used as the working electrode. Among these measurements, the applied potential was 1 V vs. Ag/AgCl. The EIS measurements were performed at the open circuit potential, ranging from 0.01 Hz to 1000 kHz. For these experiments, 0.5 mol/L KOH solution was used as the electrolyte, and a 300 W Xe lamp served as the light source.

The ORR measurements were conducted using a standard three-electrode system, with Pt and Ag/AgCl electrodes serving as the counter and reference electrodes, respectively, and the as-prepared disk-ring electrode (5.61 mm in diameter, with an electrode surface area of 0.2475 cm<sup>2</sup>) being used as the working electrode (Pine Research Instrumentation). Linear sweep voltammetry (LSV) measurements were recorded in an O<sub>2</sub>-saturated 0.5 M KOH electrolyte solution with a scan rate of 5 mV/s and a rotating speed of 1600 rpm. The potentials vs. Ag/AgCl were converted to the RHE scale by the following equation:

$$E_{RHE} = E_{Ag/AgCl} + 0.098 + 0.059 \times \text{pH}$$

In calculating the bandgap and DOS, the Geometry Optimization adopt the DFT + U approach to improve the description of strong on-site Coulomb repulsion of Ce 4 f electrons. U-J was set to 6.0 eV, which has been widely reported to be effective. The cutoff energy of 400 eV was used for all calculations, the k-point mesh of 4×4×4 was used, the energy tolerance accuracy was set to 1×10<sup>-5</sup> eV/atom, and the k-point was set to 8×8×8. The maximum force was selected at 0.03 eV/Å, and the

maximum atom displacement was placed at  $1 \times 10^{-3}$  Å. The calculations of band structure and density of states were performed using a cutoff energy of 450 eV. For the Brillouin-zone sampling, we used the  $4 \times 4 \times 4$  Monkhorst-Pack mesh, and the self-consistent convergence of the total energy is  $2.0 \times 10^{-6}$  eV/atom. The d-band center calculation is shown in the following equation:

$$\varepsilon_d = \frac{\int_{-\infty}^{\infty} n_d(\varepsilon) \varepsilon d\varepsilon}{\int_{-\infty}^{\infty} n_d(\varepsilon) d\varepsilon}$$

In calculating the electron density difference and surface  $O_V$  formation energy, the Geometry Optimization of the experimental effect of the GGA + U method with  $U = 5$  eV for the reduction of  $CeO_2$  under UV irradiation for the Ce 4f state. A model of 16.199 Å was cleaved using  $CeO_2$  (111) as a section, from which supercells were built in the range of  $2 \times 2 \times 1$ , and then a vacuum layer of 15 Å thickness was created. The cutoff energy of 400 eV was used for all calculations, the k-point mesh of  $1 \times 1 \times 1$  was used, the energy tolerance accuracy was set to  $1 \times 10^{-5}$  eV/atom, and the k-point was set to  $2 \times 2 \times 2$ . The maximum force was selected at 0.02 eV/Å, and the maximum atom displacement was placed at  $1 \times 10^{-3}$  Å. The calculations of electron density difference were performed using a cutoff energy of 400 eV, for the Brillouin-zone sampling, we used the  $4 \times 4 \times 4$  Monkhorst-Pack mesh, and the self-consistent convergence of the total energy is  $2.0 \times 10^{-6}$  eV/atom. The  $O_V$  formation energy calculation is shown in the following equation:

$$E_v^f = \frac{E_{CeO_{2-x}} - E_{CeO_2}}{x} + \frac{1}{2} E_{O_2}$$

## Characterization

Raman spectra were recorded using the Horiba (Xplora Plus) instrument. The contact angle is measured by a contact angle measuring instrument (Powereach, JC2000D). The element concentration was measured using a Thermo Scientific ICP-MS. Friction and temperature were measured using GnP POLI-500 (G&P Technology, Inc). The specific surface areas and pore size distribution were evaluated by the BET method using N<sub>2</sub> adsorption (Micromeritics, ASAP 2460). Hydrogen temperature-programmed reduction (H<sub>2</sub>-TPR) and oxygen temperature-programmed desorption (O<sub>2</sub>-TPD) measurements were performed using a BELCAT II system (MicrotracBEL, Japan). Thermal imaging is captured by Infrared thermal imager (Fluke, Fluke-Ti300). For H<sub>2</sub>-TPR, the sample was pretreated under an argon flow (30 mL/min) by heating from room temperature to 300 °C at a rate of 10 °C/min and holding for 60 min. After cooling to 50 °C, a 10% H<sub>2</sub>/Ar gas mixture (50 mL/min) was introduced for 0.5 h. Once the baseline stabilized, the temperature was increased to 800 °C at a rate of 10 °C/min under a 10% H<sub>2</sub>/Ar flow (30 mL/min). For O<sub>2</sub>-TPD, the sample was similarly pretreated in helium at 300 °C for 1 h. After cooling and stabilization, a 10% O<sub>2</sub>/Ar gas mixture (50 mL/min) was introduced for 0.5 h. The sample was then heated to 800 °C at 10 °C/min under a 10% O<sub>2</sub>/Ar flow (30 mL/min). Current–voltage (I–V) characteristics were measured using an Agilent B1500A Semiconductor Device Analyzer in conjunction with an EverBeing EB-8 probe station.

The X-ray Photoelectron Spectroscopy (XPS) instrument utilized in this research was the ESCALAB250Xi model, manufactured by Thermo Scientific. After testing, the XPS spectra were analyzed using CasaXPS software and calibrated based on the

149 C1s peak (284.6 eV). The amount of  $Ce^{3+}$  was calculated based on the Ce 3d XPS  
150 spectra as the following equation:

$$p(Ce^{3+}) = \frac{Ce^{3+}}{Ce^{4+} + Ce^{3+}} \times 100\%$$

151  
152 The density of  $O_v$  was calculated based on the O 1S XPS spectra as the following  
153 equation:

$$p(O_v) = \frac{O_v}{O_v + O_L + O_C} \times 100\%$$

154  
155 The diffuse reflection spectra were measured by UV-Vis spectrophotometer  
156 (PerkinElmer, Lambda 1050+). The bandgap energy ( $E_g$ ) was calculated as the  
157 following formula:

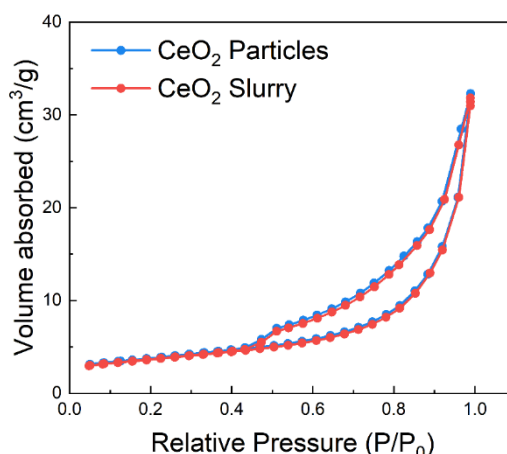
$$\alpha h\nu = A(h\nu - E_g)^n$$

158  
159 where A, h,  $\nu$ , and  $\alpha$  are constant values that refer to a constant, Plank  
160 constant, light frequency and absorption coefficient, respectively.  
161

## 162 Calculation details

163 The PWmat program with GPU acceleration was employed to  
164 investigate the peak-to-valley difference of the Si (100) surface based on  
165 density functional theory (DFT). The exchange-correlation interactions  
166 were treated using the Perdew-Burke-Ernzerhof (PBE) functional within  
167 the generalized gradient approximation (GGA). Norm-conserving  
168 pseudopotentials were utilized, and the plane-wave energy cutoff was set  
169 to 544 eV. The surface model consisted of three atomic layers and a  
170 vacuum region of 15 Å. Structural optimization was performed with  
171 convergence criteria of forces less than 0.1 eV Å<sup>-1</sup> and energy changes  
172 below 10<sup>-4</sup> eV. Converged energy:  $E = -1.362572371481 \times 10^4$  eV.

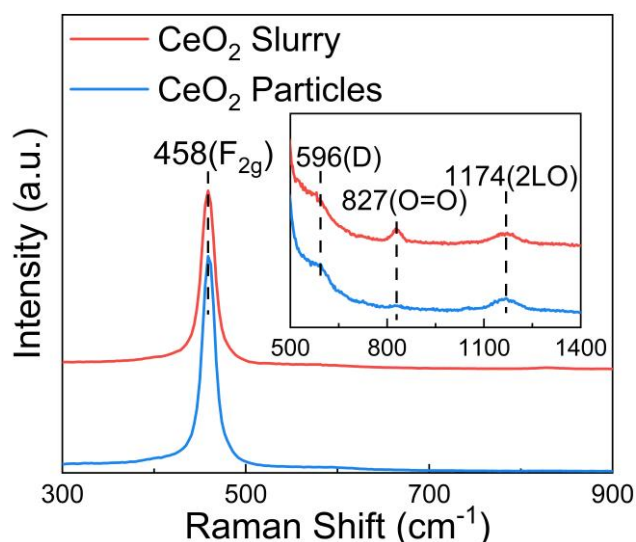
## 173    **Supplementary Figures**



174    **Supplementary Fig. S1.** BET surface area of CeO<sub>2</sub> particles and CeO<sub>2</sub> slurry.

175

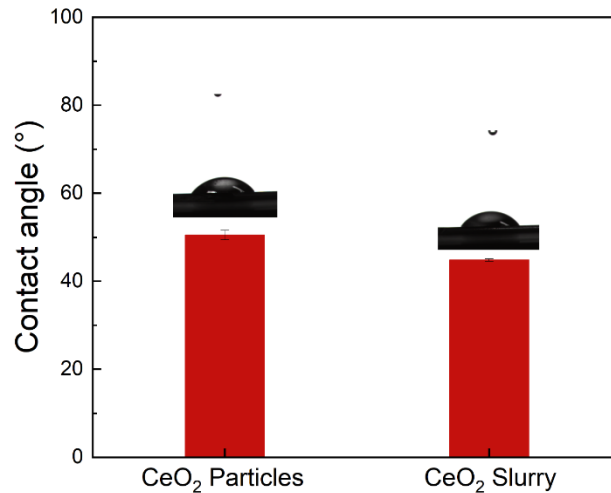
176    **Discussion:** The specific surface areas of CeO<sub>2</sub> particles and CeO<sub>2</sub> slurry  
177    were measured using the Brunauer–Emmett–Teller (BET). The specific  
178    surface area of CeO<sub>2</sub> particles was 13.26 m<sup>2</sup>/g, whereas that of the CeO<sub>2</sub>  
179    slurry was slightly lower, at 12.92 m<sup>2</sup>/g. This minor reduction indicates  
180    that the slurry preparation process has a negligible impact on the overall  
181    surface area of the CeO<sub>2</sub> material. Additionally, the nitrogen  
182    adsorption-desorption isotherms for both samples exhibited a typical type  
183    IV profile with a prominent hysteresis loop characteristic of mesoporous  
184    structures.



**Supplementary Fig. S2.** Raman spectrum, inset: the 500-1300 cm<sup>-1</sup> shift region.

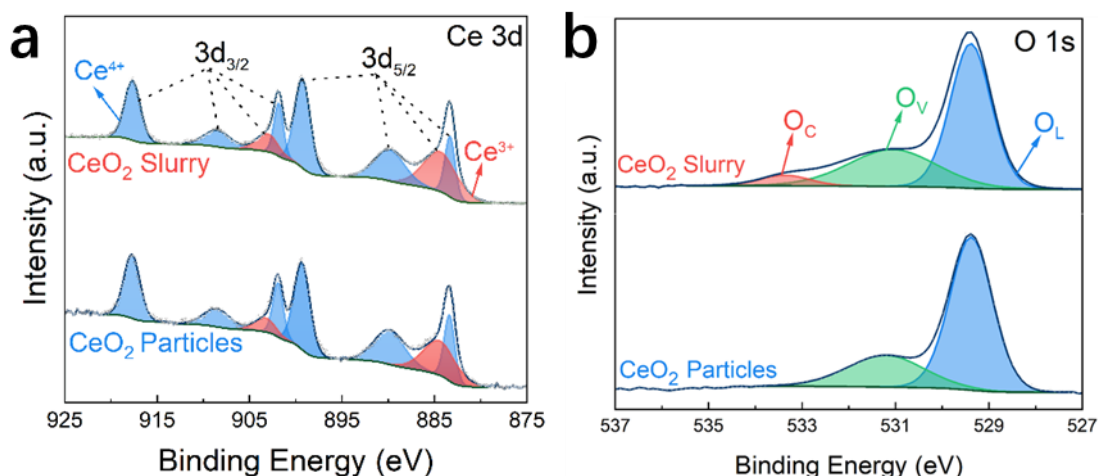
**Discussion:** The CeO<sub>2</sub> particles were primarily characterized by a peak at 458 cm<sup>-1</sup>, corresponding to the F<sub>2g</sub> mode of the CeO<sub>2</sub> lattice<sup>1</sup>. Additional peaks at 596 cm<sup>-1</sup>, 827 cm<sup>-1</sup>, and 1174 cm<sup>-1</sup> were associated with defect-induced modes (D), adsorbed peroxo/superoxo species (O–O stretching), and longitudinal optical (2LO)<sup>2</sup>. Adsorbed O<sub>2</sub><sup>2-</sup>/O<sub>2</sub><sup>-</sup> species act as intermediates in the re-oxidation of reduced CeO<sub>2-x</sub> to CeO<sub>2</sub><sup>3</sup>. Electrons donated by Ce<sup>3+</sup> cations drive their conversion to Ce<sup>3+</sup>.

The ratio of the F<sub>2g</sub> to defect peak intensities ( $I_{F_{2g}} / I_D$ ) serves as a sensitive measure of lattice order versus defectiveness. A higher ratio indicates better crystallinity and fewer defects, while a lower ratio reflects increased lattice disorder from oxygen vacancies, dopant effects, or other imperfections. In Fig. S2, these ratios are 44.16 for CeO<sub>2</sub> particles and 38.64 for CeO<sub>2</sub> slurry, confirming a higher defect concentration in the slurry.



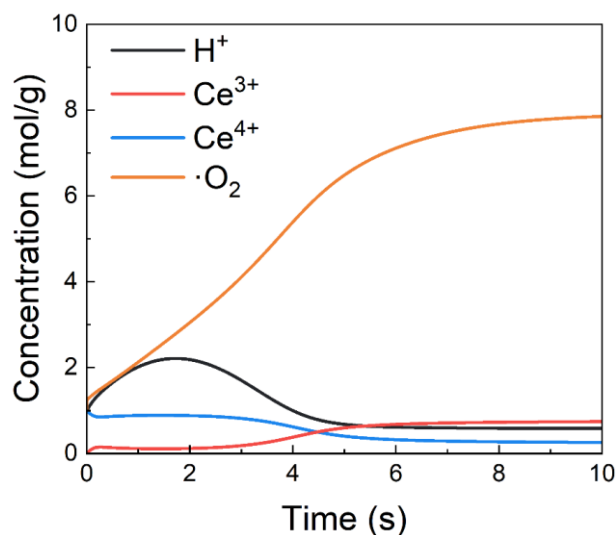
**Supplementary Fig. S3.** Contact angle of CeO<sub>2</sub> particles and CeO<sub>2</sub> slurry layer on Si wafers surface with deionized water.

**Discussion:** Introducing oxygen vacancies leads to enhanced surface charge accumulation, which reduces particle agglomeration and accounts for the observed decrease in zeta potential. Moreover, oxygen vacancies increase the surface free energy, promoting more effective interactions between the CeO<sub>2</sub> abrasive and Si wafers. This effect is supported by contact angle measurements, where the CeO<sub>2</sub> slurry demonstrated a reduced contact angle of 44.56°, representing a decrease of 5.93° compared to CeO<sub>2</sub> particles. This reduction is attributed to the increased hydrophilicity resulting from water dissociative adsorption on the CeO<sub>2</sub> surface following the introduction of oxygen vacancies.



**Supplementary Fig. S4. a Ce 3d, b O 1s XPS spectra of CeO<sub>2</sub> particles and CeO<sub>2</sub> slurry.**

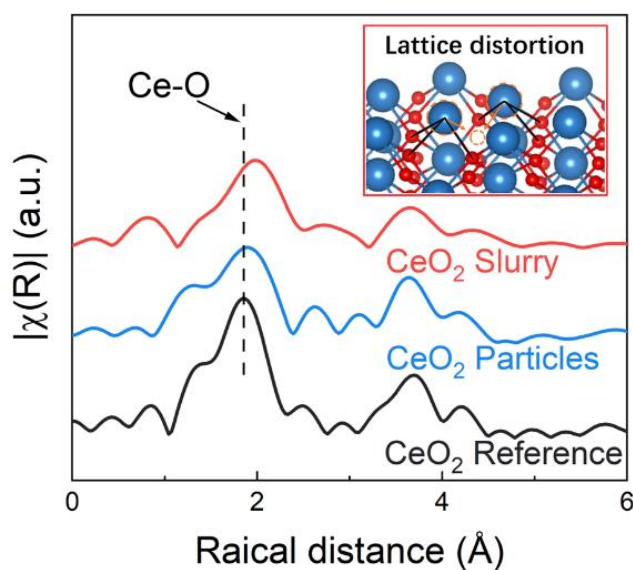
**Discussion:** The Ce 3d spectra (Fig. S4a) comprise five spin-orbit split double peaks (3d<sub>3/2</sub>, 3d<sub>5/2</sub>), with 903.4 eV and 885.2 eV corresponding to Ce<sup>3+</sup> signals<sup>4</sup>. Combined with the O 1s spectra (Fig. S4b), the results indicate that Janus CeO<sub>2</sub> slurry has a higher concentration of active sites. The relative contents of Ce<sup>3+</sup> and oxygen vacancies are summarized in Table S1. The O 1s XPS spectrum shows peaks at 531.17 eV and 529.37 eV, which are attributed to oxygen vacancies (O<sub>V</sub>) and lattice oxygen (O<sub>L</sub>), respectively, while the peak at 533.36 eV corresponds to chemisorbed oxygen species (O<sub>C</sub>). Calculations indicate that the O<sub>V</sub> density is 30.17% in CeO<sub>2</sub> particles and increases to 34.02% in the CeO<sub>2</sub> slurry. The O<sub>C</sub> originate from adsorbed peroxides (O<sub>2</sub><sup>2-</sup>/O<sub>2</sub><sup>-</sup>) on the CeO<sub>2</sub> surface.



**Supplementary Fig. S5.** Disproportionation reaction of CeO<sub>2</sub> with H<sub>2</sub>O<sub>2</sub> under alkaline conditions.

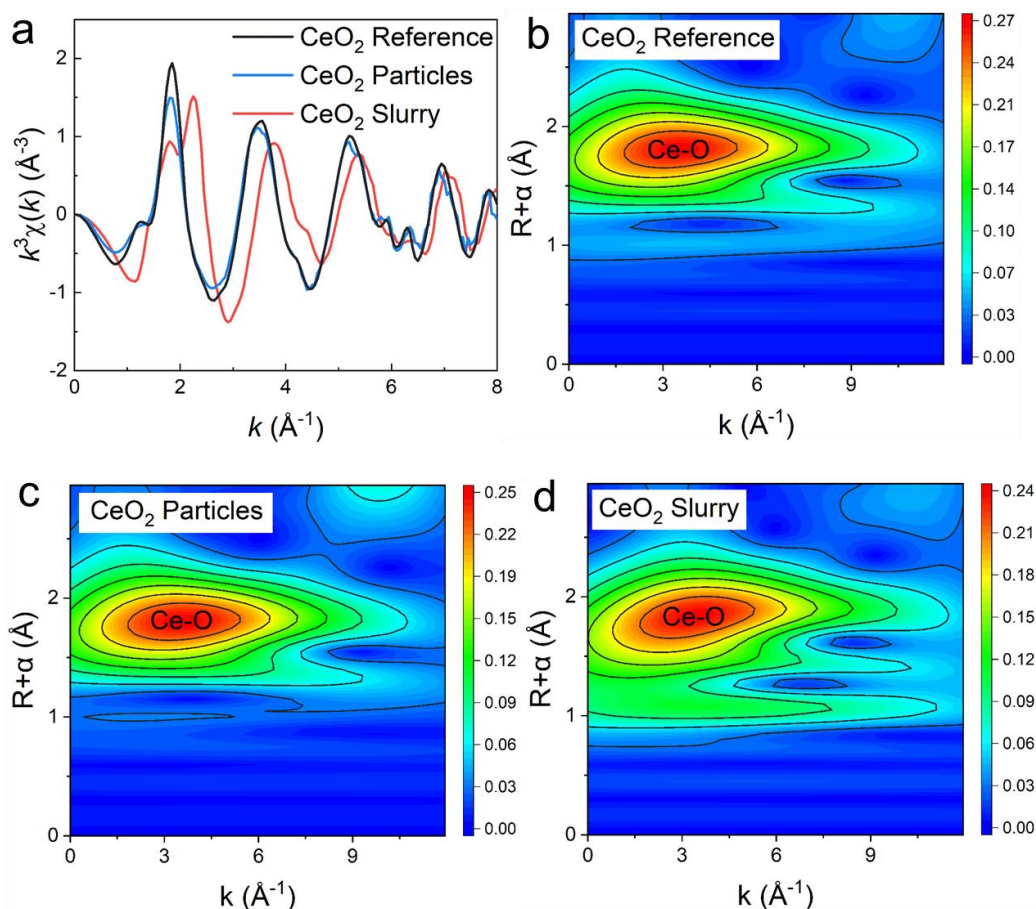
**Discussion:** During the photocatalytic CMP process, oxygen vacancy concentration of CeO<sub>2</sub> increases when CeO<sub>2</sub> is combined with H<sub>2</sub>O<sub>2</sub>. Oxygen vacancies migrate to energetically favorable lattice positions, and Ce<sup>3+</sup> relocates to the crystal surface. Due to the increased ionic radius, they are better accommodated spatially<sup>5</sup>. For this mechanism, a reasonable model for the superoxide disproportionation reaction mechanism at the cerium dioxide nanoparticle surface<sup>6</sup>.

The CeO<sub>2</sub> slurry shows alkalinity. Therefore, the Runge-Kutta method was used for modeling. Under alkaline conditions, the conversion between Ce<sup>3+</sup> and Ce<sup>4+</sup> occurs and eventually reaches equilibrium. When hydrogen peroxide is added, Ce<sup>4+</sup> actively controls the superoxide, resulting in a minor transformation of Ce<sup>3+</sup> ions, leading to oxygen vacancies.



**Supplementary Fig. S6.** Radial distribution function from EXAFS, inset: lattice distortion and coordination environment change in CeO<sub>2</sub> slurry.

**Discussion:** The R-space spectra and fitting data (Table S2) further reveal variations in the Ce-O bond length within the CeO<sub>2</sub> slurry. Introducing oxygen vacancies results in the elongation of first-shell Ce-O bonds, leading to surface lattice distortion. These findings are further supported by the K-space EXAFS fitting spectra and wavelet transform analysis.

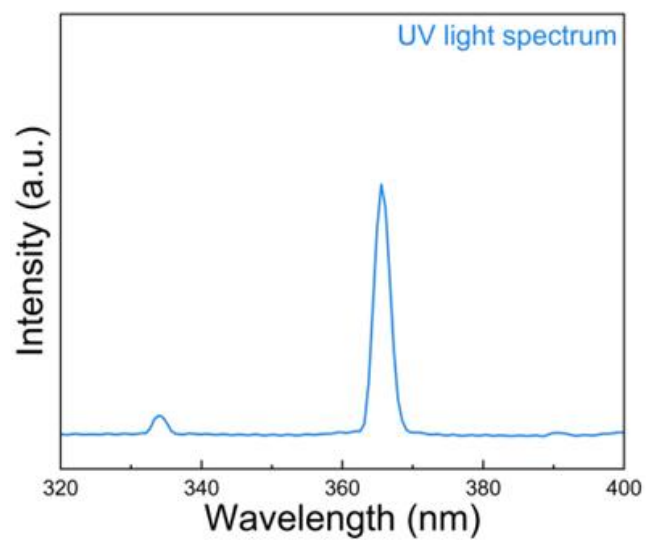


**Supplementary Fig. S7.** **a** The corresponding K space EXAFS fitting spectra of CeO<sub>2</sub> reference, CeO<sub>2</sub> particles, and CeO<sub>2</sub> slurry. Wavelet transform of Ce K-edge EXAFS spectra for **b** CeO<sub>2</sub> reference, **c** CeO<sub>2</sub> particles, and **d** CeO<sub>2</sub> slurry.

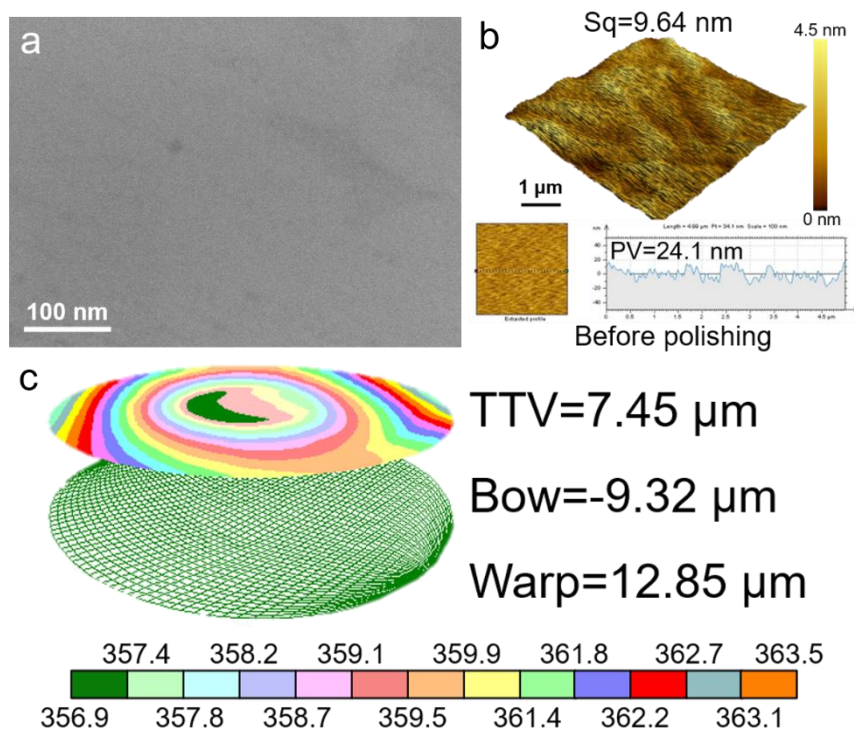
**Discussion:** Combining the results of the EXAFS fits (Table S2) reveals a lower coordination number and a higher Debye-Waller factor for the CeO<sub>2</sub> slurry, suggesting a more disordered surface structure. Additionally, oxygen vacancies partially contribute to the Ce-O-Ce shell, thereby promoting greater electron delocalization.

The local chemical environment and physical structure around the Ce atoms were investigated using Ce K-edge EXAFS spectra. From the wavelet transform contour plots (Fig. S7a), each sample exhibited distinct maxima attributed to the scattering effect of neighboring O atoms (Ce-O bonding, first shell,  $k \sim 3.6 \text{ \AA}^{-1}$ ). Compared to the CeO<sub>2</sub> reference and

267 CeO<sub>2</sub> particles, the intensity of Ce-O bonding was significantly reduced  
268 for the CeO<sub>2</sub> slurry, indicating a higher concentration of oxygen  
269 vacancies. This reduction is attributed to homogeneous interfaces that  
270 disrupt the Ce atom arrays, inducing lattice distortions and suppressing  
271 multiple scattering of photoelectrons. Correspondingly, the Ce-O  
272 coordination numbers quantitatively support these findings.

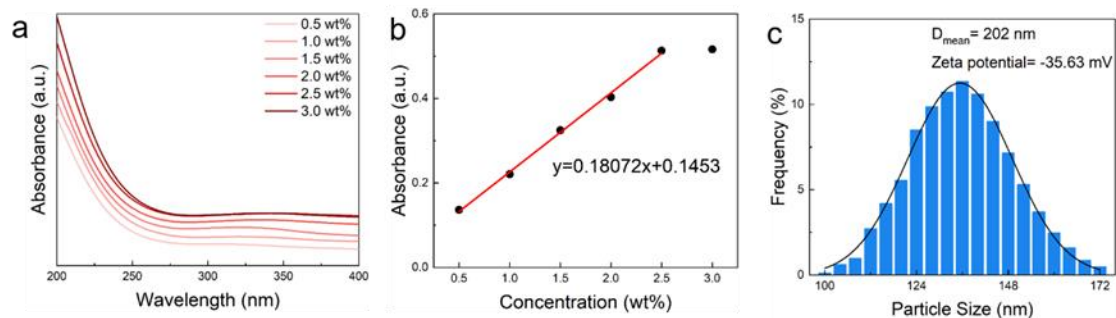


273 **Supplementary Fig. S8.** Emission spectrum of the UV light source centered at 365  
274 nm.



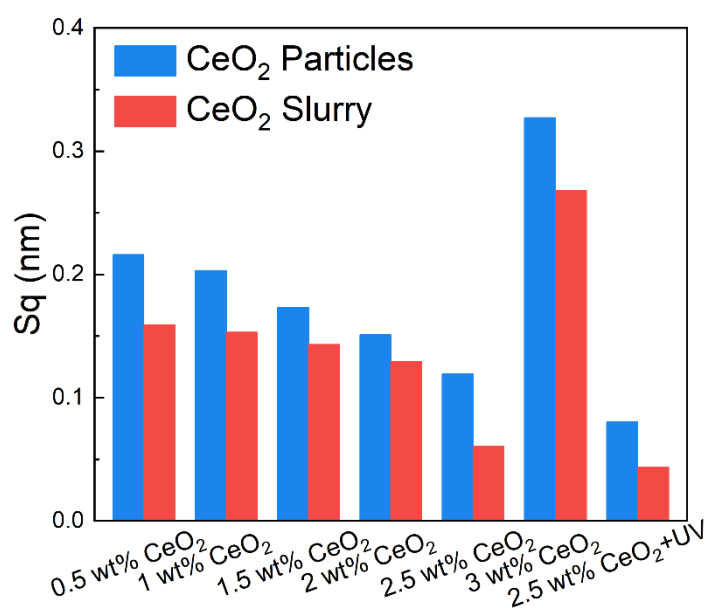
275 **Supplementary Fig. S9.** **a** 3D AFM image and height profiles. **b** SEM image. **c**

276 Geometric parameter image of Si CMP using CeO<sub>2</sub> particles.

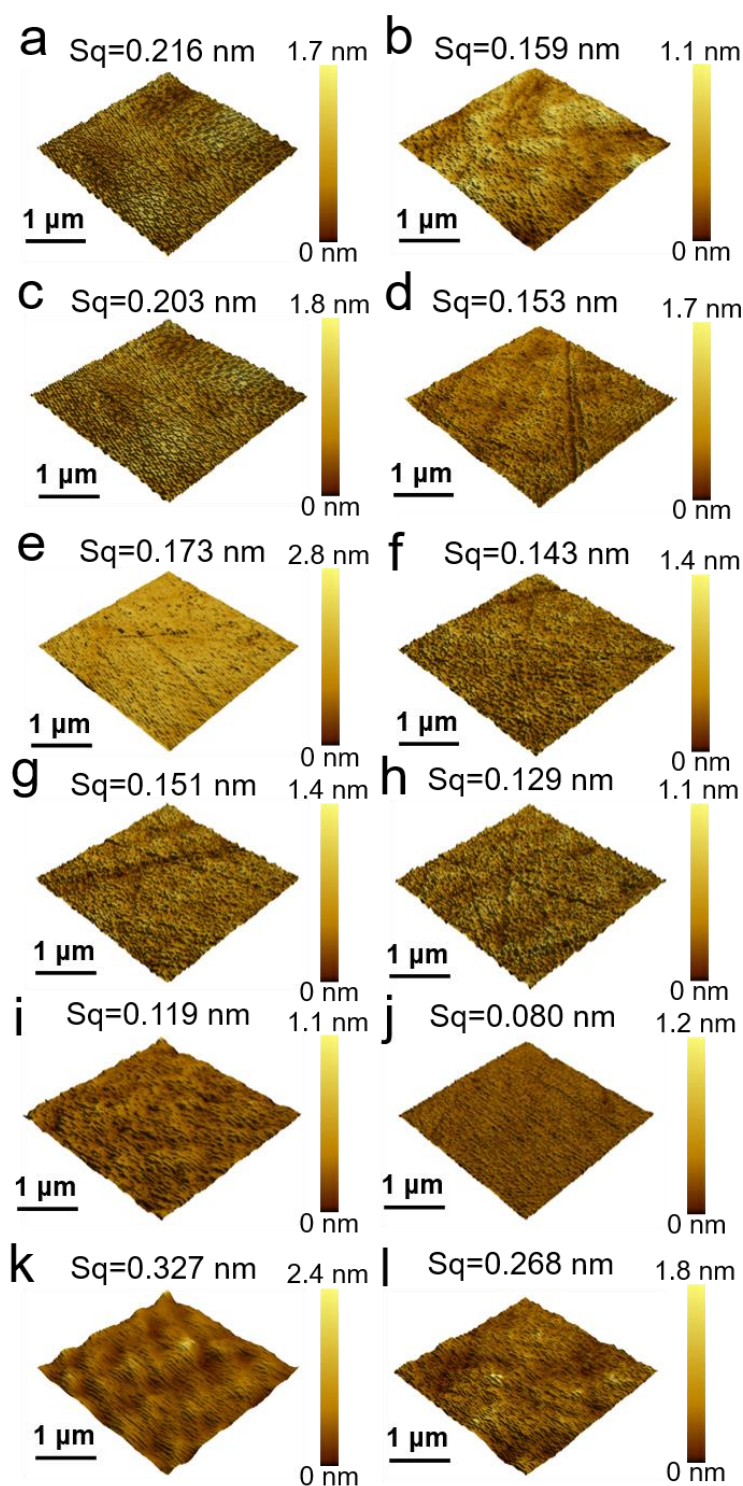


**Supplementary Fig. S10.** (a) UV-Vis absorption spectra of CeO<sub>2</sub> slurry of different concentrations. (b) This fit shows a linear trend with a slope of 0.18072 called the exponent coefficient. (c) Particle size distribution histogram and zeta potential of 3 wt.% CeO<sub>2</sub>, which exhibits an average CeO<sub>2</sub> particle size of 136.5 nm and zeta potential of -35.63 mV.

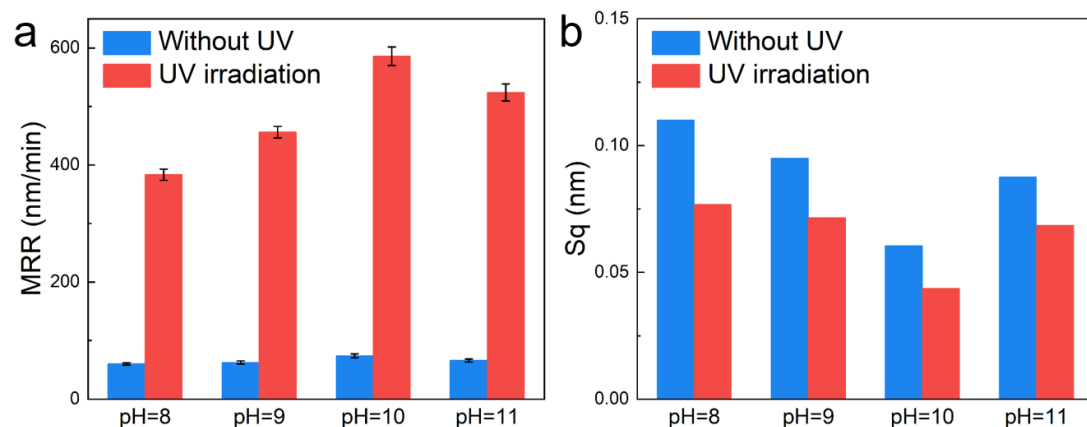
**Discussion:** The intensity of the characteristic absorption peak gradually increased, with the CeO<sub>2</sub> slurry reaching maximum absorbance at a concentration of 2.5 wt.%, indicating that the CeO<sub>2</sub> abrasives were approaching saturation in the dispersion. However, upon further increasing the CeO<sub>2</sub> concentration, the abrasives exceeded saturation, resulting in particle aggregation and sedimentation. This supersaturation led to a decrease in both the dispersion quality and the stability of the polishing solution.



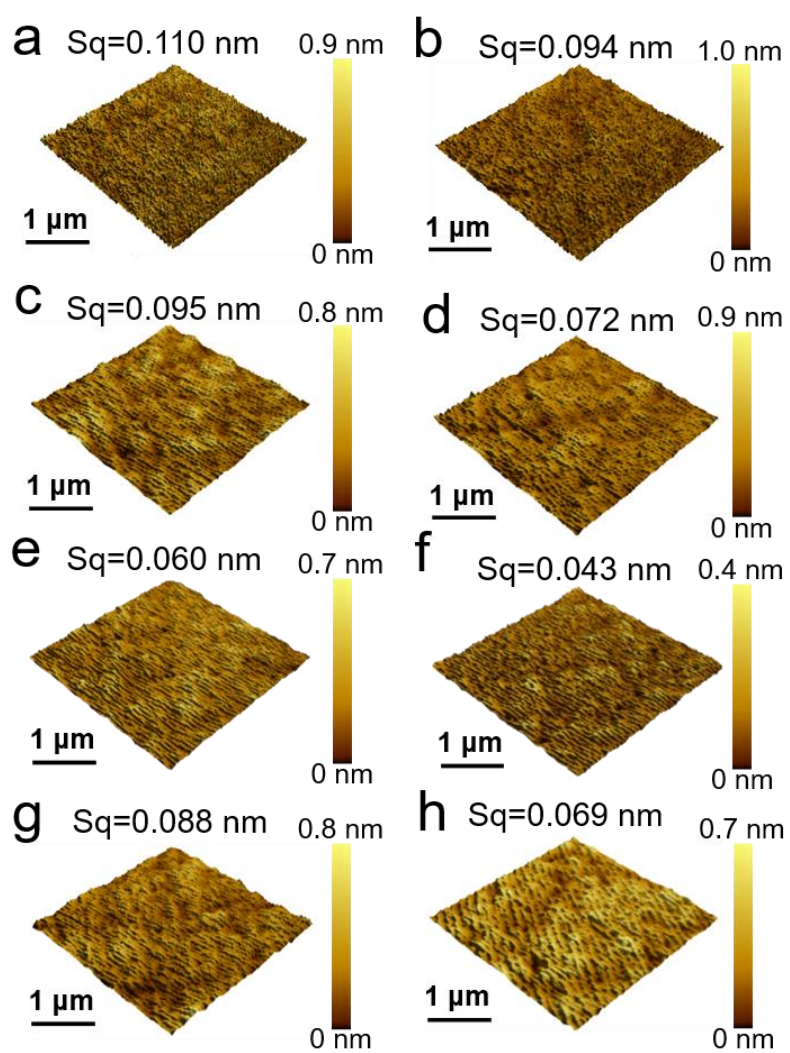
291 **Supplementary Fig. S11.** Sq comparison for CeO<sub>2</sub> particles and Janus CeO<sub>2</sub> slurry at  
 292 various weight concentrations and UV illumination.



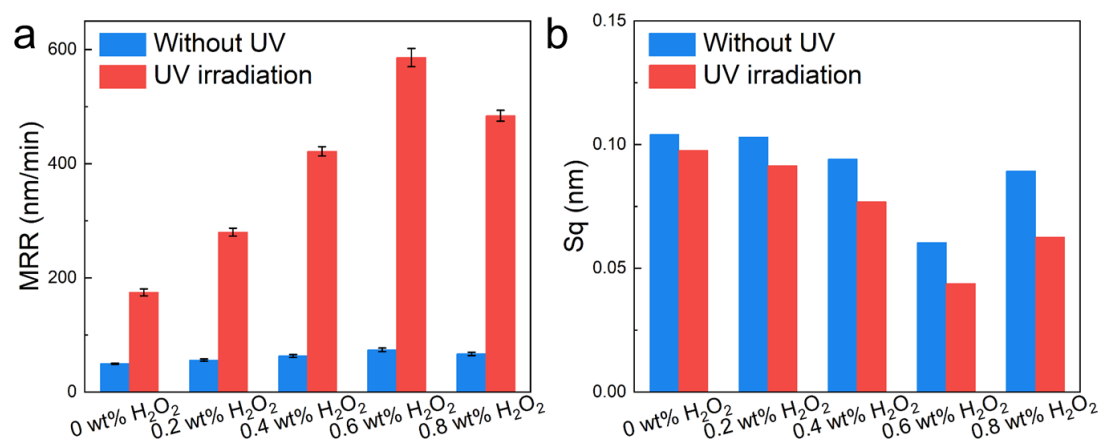
293 **Supplementary Fig. S12.** 3D AFM images of Si wafer surfaces polished under  
 294 various weight concentrations CeO<sub>2</sub> particles and Janus CeO<sub>2</sub> slurry: **a** 0.5 wt%  
 295 (particles); **b** 0.5 wt% (slurry); **c** 1 wt% (particles); **d** 1 wt% (slurry); **e** 1.5 wt%  
 296 (particles); **f** 1.5 wt% (slurry); **g** 2 wt% (particles); **h** 2 wt% (slurry); **i** 2.5 wt%  
 297 (particles); **j** 2.5 wt% (particles)+UV; **k** 3 wt% (particles); **l** 3 wt% (slurry).



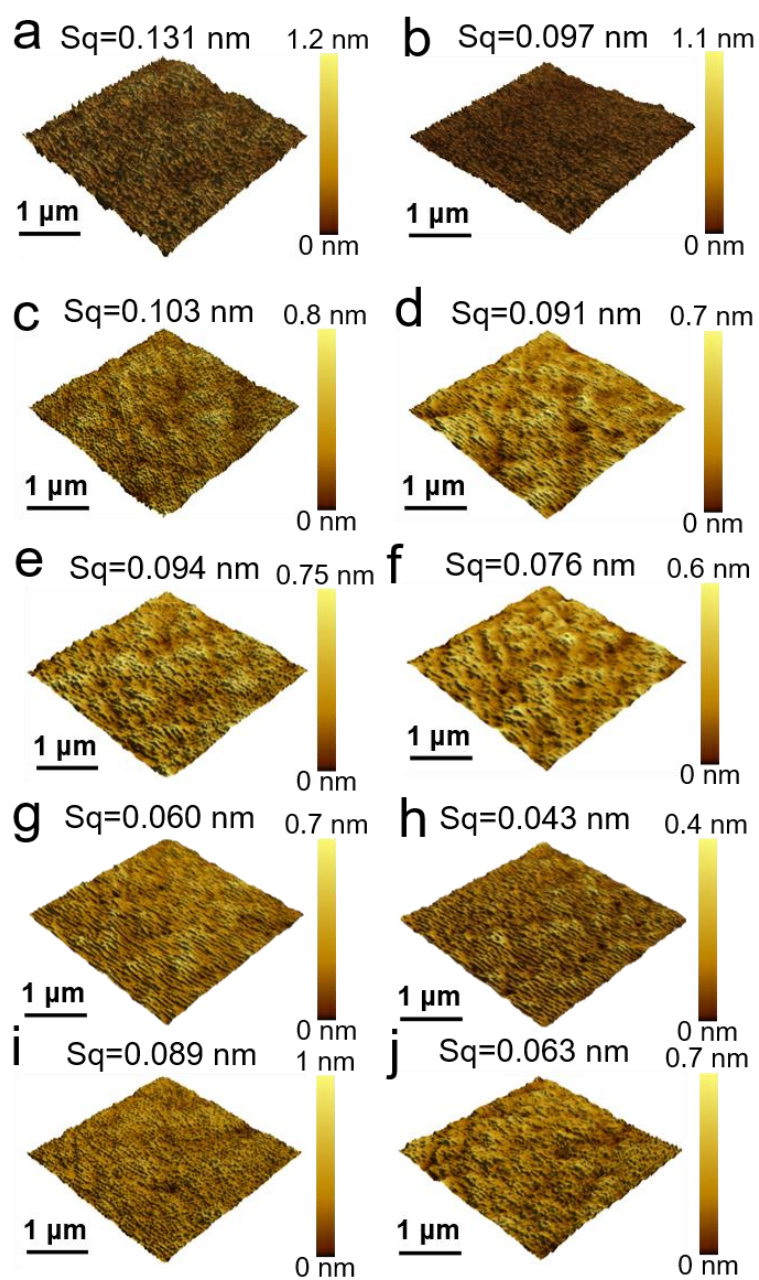
298 **Supplementary Fig. S13.** Comparison of **a** MRR, **b** Sq for Si wafers polished at  
 299 different pH levels.



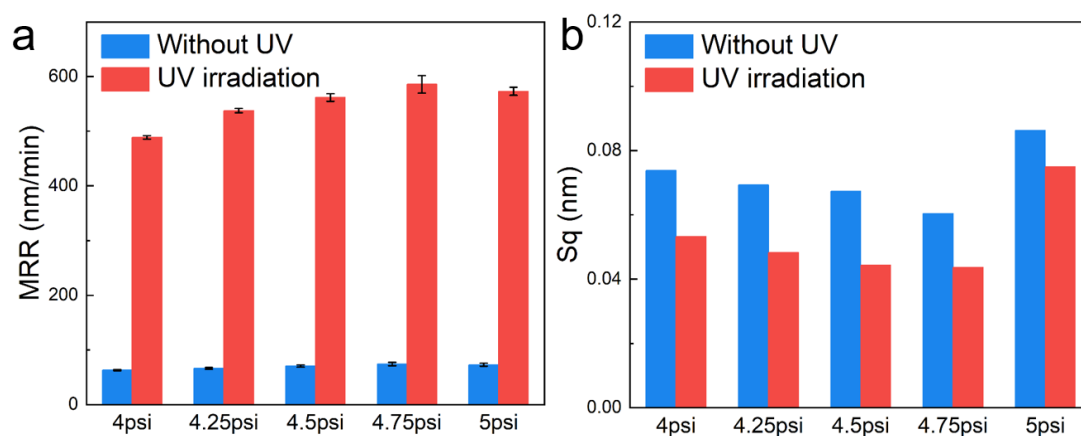
300 **Supplementary Fig. S14.** 3D AFM images of Si wafer surfaces polished under  
 301 different pH: **a** pH=8; **b** pH=8 (UV); **c** pH=9; **d** pH=9 (UV); **e** pH=10; **f** pH=10 (UV);  
 302 **g** pH=11; **h** pH=11 (UV).



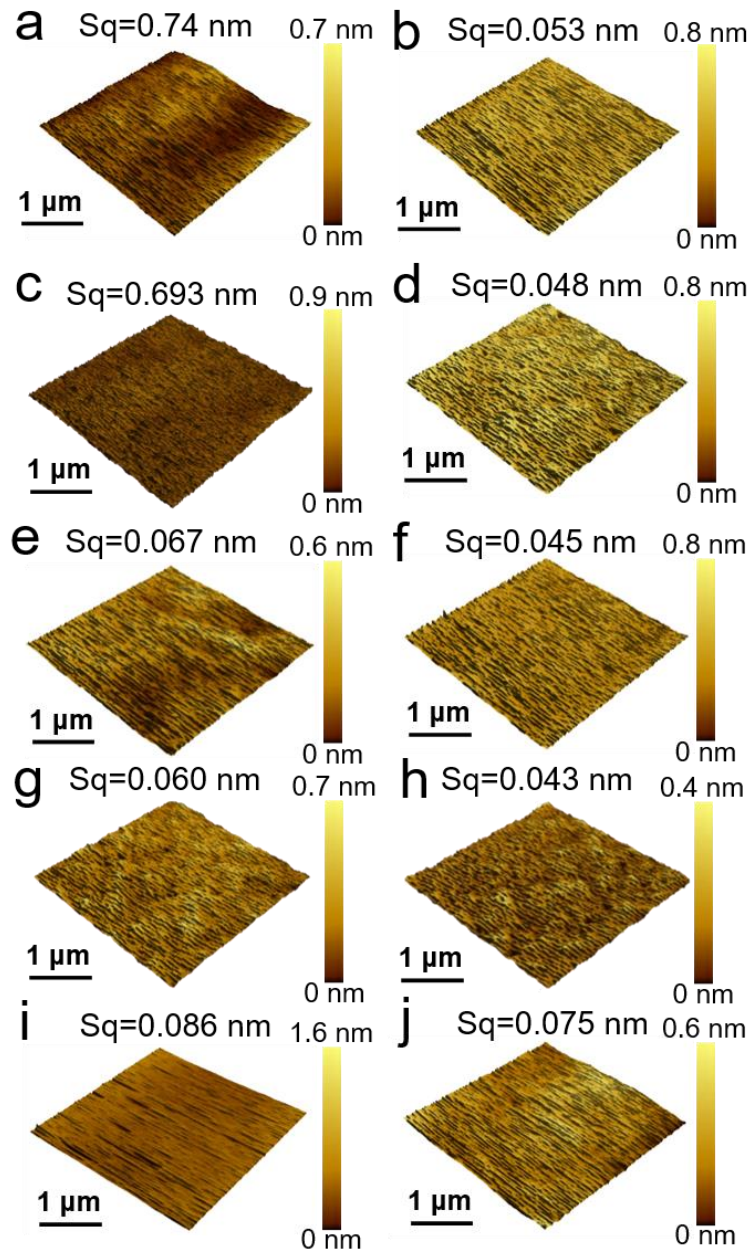
303 **Supplementary Fig. S15.** Comparison of **a** MRR, **b** Sq for the Si wafers polished  
 304 under different weight concentrations of  $H_2O_2$ .



305 **Supplementary Fig. S16.** 3D AFM images of Si wafer surfaces polished under  
 306 different weight concentrations of  $H_2O_2$ : **a** 0 wt%; **b** 0 wt% (UV); **c** 0.2 wt%; **d** 0.2  
 307 wt% (UV); **e** 0.4 wt%; **f** 0.4 wt% (UV); **g** 0.6 wt%; **h** 0.6 wt% (UV); **i** 0.8 wt%; **j** 0.8  
 308 wt% (UV).

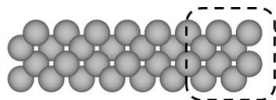


309 **Supplementary Fig. S17.** Comparison of **a** MRR, **b** Sq on the Si wafers polishing  
 310 under different polishing pressure.

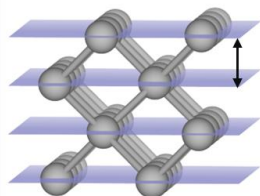


311 **Supplementary Fig. S18.** 3D AFM images of Si wafer surfaces polished under  
 312 different polishing pressure: **a** 4 psi; **b** 4 psi (UV); **c** 4.25 psi; **d** 4.25 psi (UV); **e** 4.5  
 313 psi; **f** 4.5 psi (UV); **g** 4.75 psi; **h** 4.75 psi (UV); **i** 5 psi; **j** 5 psi (UV).

Atomic arrangement of  
the Si (100) surface

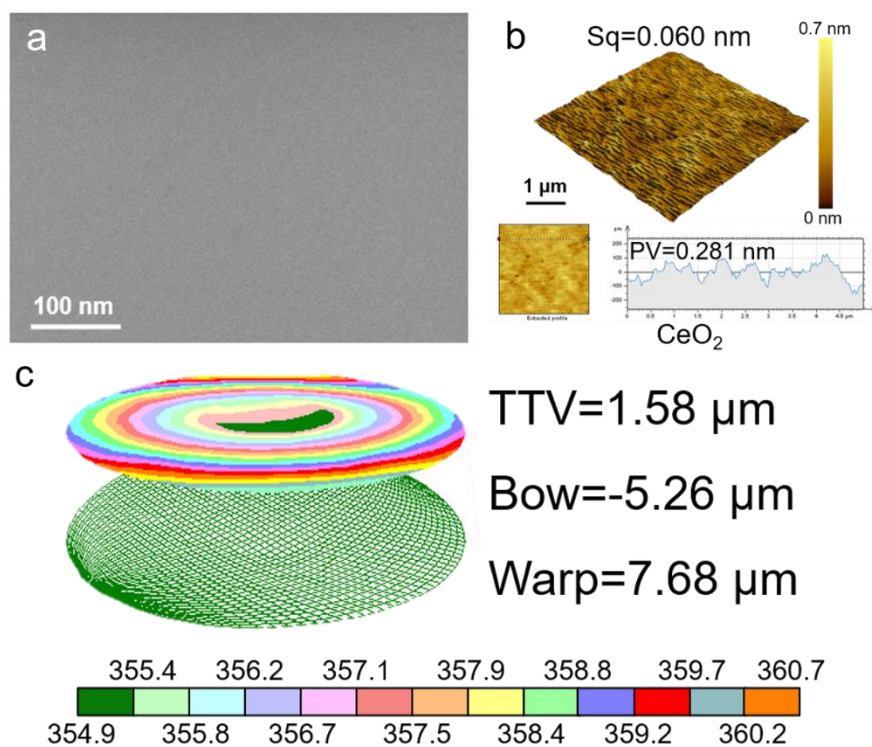


Side view of the Si (100) surface atoms

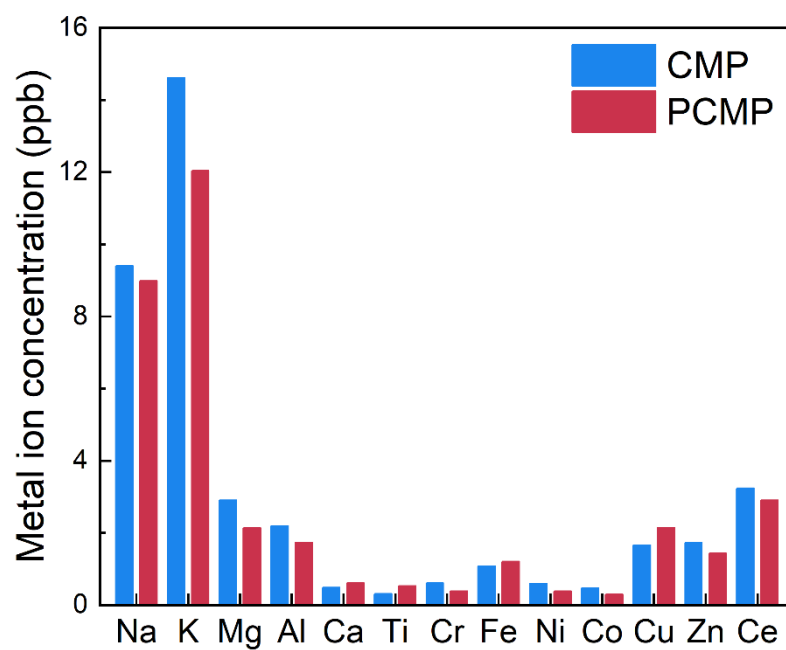


Atomic layer spacing: 1.36 Å

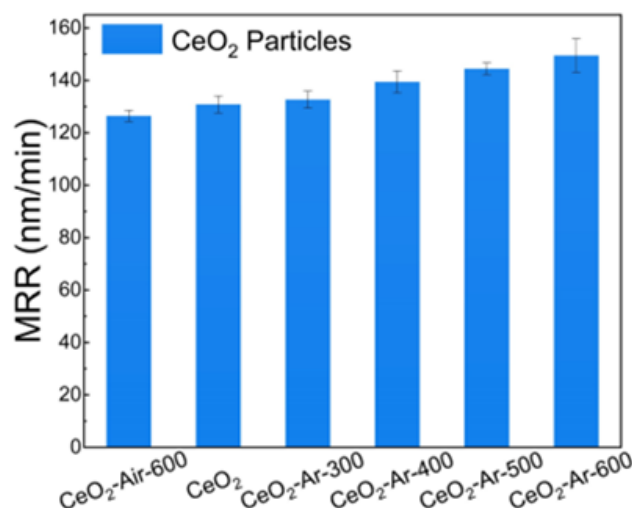
314 **Supplementary Fig. S19.** Atomic arrangement of the Si (100) surface.



315 **Supplementary Fig. S20. a** 3D AFM image and height profiles. **b** SEM image. **c** Si  
 316 Geometric parameter image using Janus CeO<sub>2</sub> slurry (2.5 wt%).



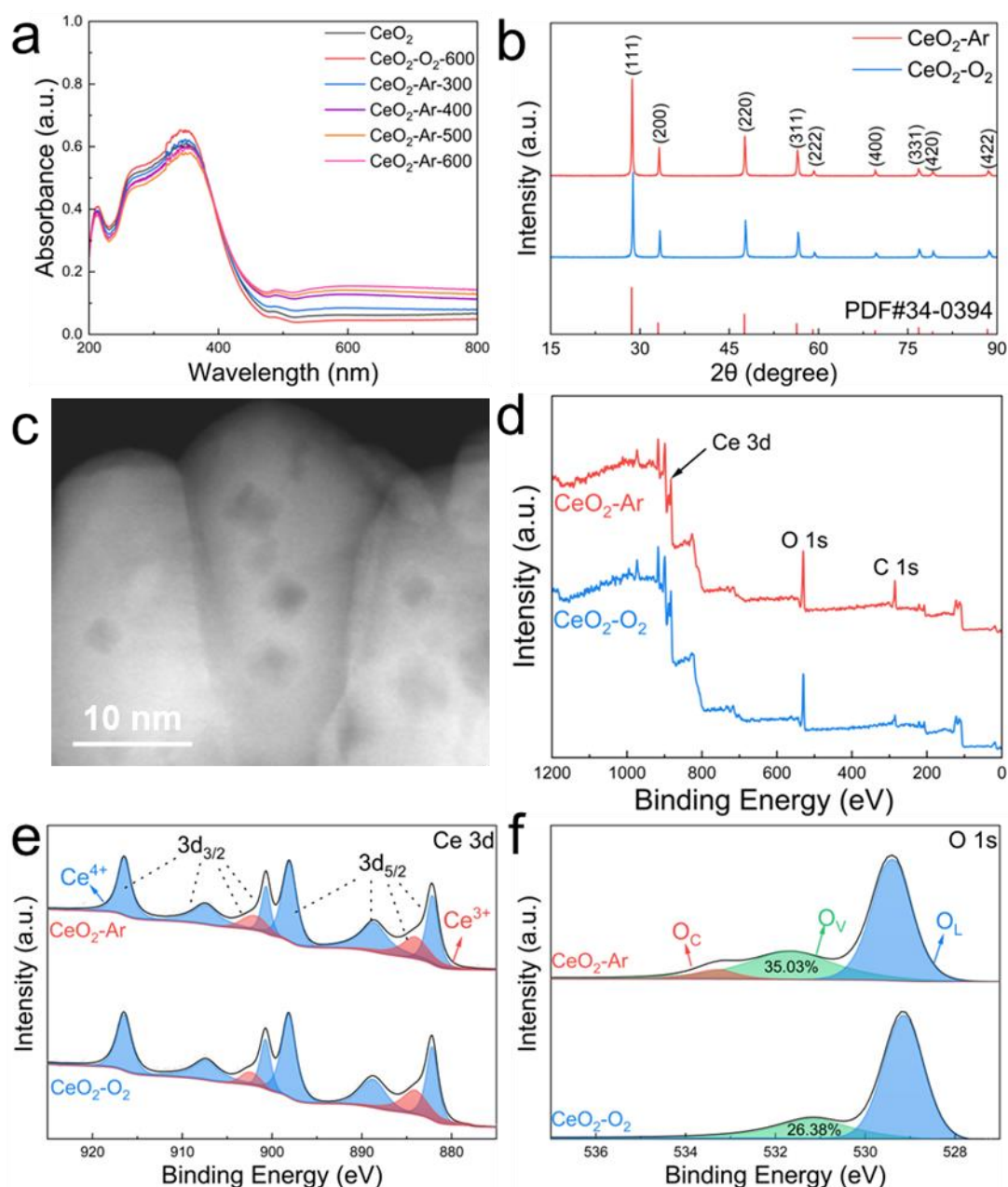
317 **Supplementary Fig. S21.** The polished Si wafers of metal element concentration.



**Supplementary Fig. S22. a** Comparison of MRR on SiO<sub>2</sub> wafers with CeO<sub>2</sub> under different conditions.

**Discussion:** The MRR of SiO<sub>2</sub> during CMP using CeO<sub>2</sub> particles subjected to different calcination conditions was investigated. Samples calcined in air (CeO<sub>2</sub>-Air-600) exhibited the lowest MRR, attributed to the reduced concentration of oxygen vacancies, as air calcination tends to replenish lattice oxygen. In contrast, CeO<sub>2</sub> samples calcined in an Ar atmosphere showed progressively higher MRRs with increasing calcination temperatures (300–600 °C), due to the enhanced formation of oxygen vacancies. These results indicate that oxygen vacancies in CeO<sub>2</sub> can improve its chemical reactivity, thereby promoting more efficient SiO<sub>2</sub> removal during CMP.

However, the formation of oxygen vacancies reaches a saturation point beyond which further increase becomes limited. Since the oxidation of the Si surface is the rate-determining step in the CMP process, boosting the oxidation rate can result in several-fold increases in MRR. In contrast, increasing oxygen vacancy concentration alone yields only marginal improvements.



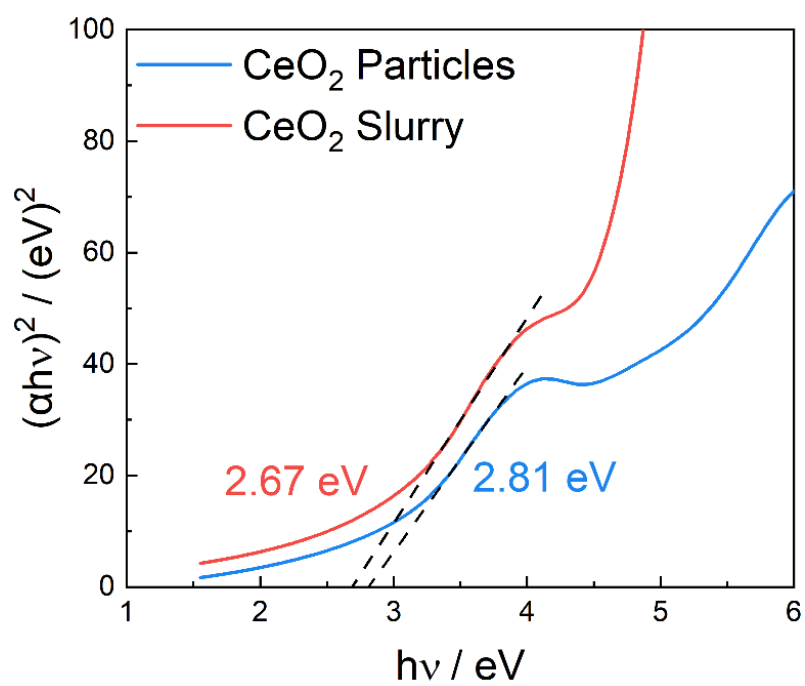
**Supplementary Fig. S23.** **a** UV-Vis spectra. **b** XRD pattern. **c** HAADF-STEM of  $\text{CeO}_2\text{-Ar-600}$ . **d** XPS spectra, **e** Ce 3d, **d** O 1s XPS spectra of  $\text{CeO}_2\text{-Ar}$  and  $\text{CeO}_2\text{-O}_2$ .

**Discussion:** Oxygen treatment tends to fill oxygen vacancies, while annealing in Ar atmosphere promotes oxygen diffusion out of the lattice, thereby generating oxygen vacancies. As shown in Fig. S23a, visible light absorption in the 500–800 nm range gradually increases with the

concentration of oxygen vacancies, suggesting enhanced defect-related electronic transitions.

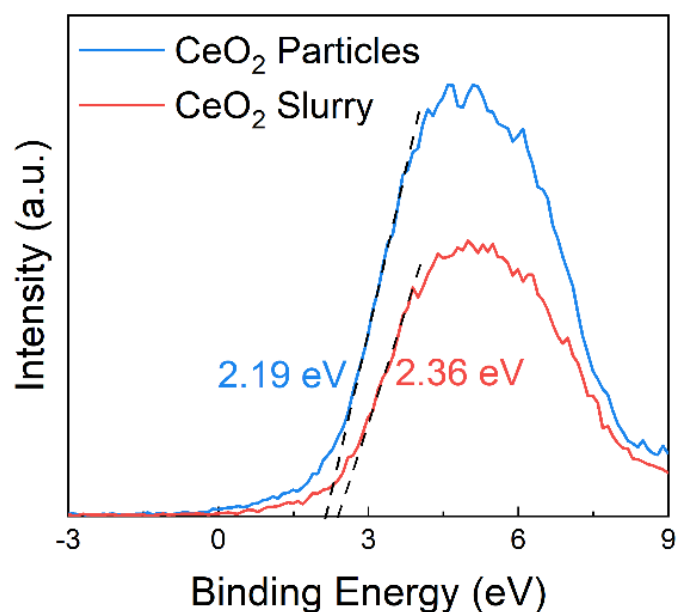
XRD analysis (Fig. S23b) reveals a leftward shift of diffraction peaks in CeO<sub>2</sub> samples annealed under Ar (CeO<sub>2</sub>-Ar) compared to those treated in O<sub>2</sub> (CeO<sub>2</sub>-O<sub>2</sub>), consistent with lattice expansion. This shift is attributed to two coupled effects: (1) the formation of oxygen vacancies, which locally disrupt the fluorite structure, and (2) the partial reduction of Ce<sup>4+</sup> to Ce<sup>3+</sup>, whose larger ionic radius (1.14 Å for Ce<sup>3+</sup> vs. 0.97 Å for Ce<sup>4+</sup> in 8-fold coordination) leads to elongation of Ce-O bonds to preserve charge neutrality<sup>7</sup>.

These structural modifications are further corroborated by HAADF-STEM and XPS (Fig. S23c-f). The HAADF-STEM image clearly displays abundant surface defects on CeO<sub>2</sub>-Ar particles. XPS analysis of the O 1s spectra shows that the oxygen vacancy contribution increases from 26.38% in CeO<sub>2</sub>-O<sub>2</sub> to 35.03% in CeO<sub>2</sub>-Ar (Fig. S23f), validating the effectiveness of Ar annealing in enhancing defect concentration. Together, these results demonstrate a strong correlation between reduction state, lattice parameter, and oxygen vacancy concentration, highlighting the critical role of defect thermodynamics in tailoring the structural and electronic properties of ceria-based materials.



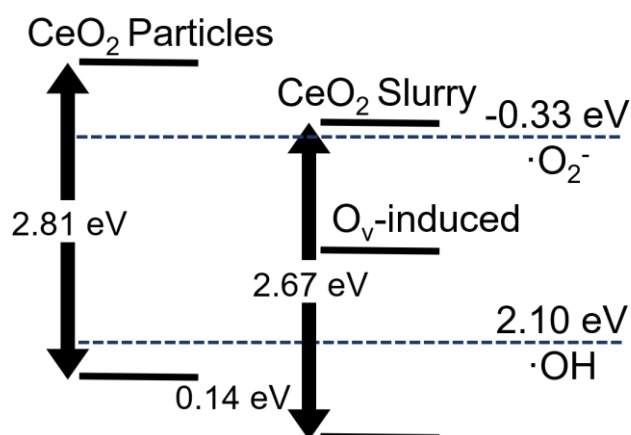
364

**Supplementary Fig. S24.** Tauc plot of CeO<sub>2</sub> particles and CeO<sub>2</sub> slurry.



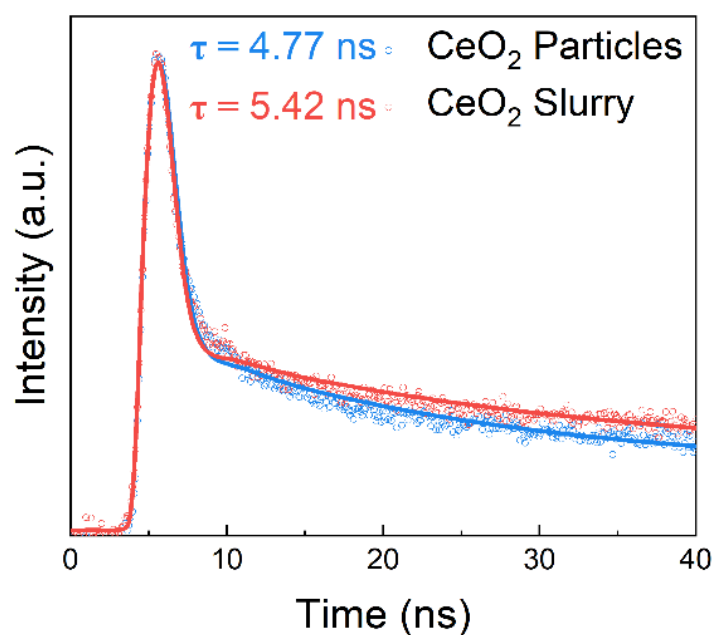
**Supplementary Fig. S25.** UPS valence band spectra of CeO<sub>2</sub> particals and slurry.

**Discussion:** The presence of oxygen vacancies causes electrons at the top of the valence band to shift to lower energy levels<sup>8</sup>, resulting in valence band potentials of 2.36 eV for CeO<sub>2</sub> particles and 2.19 eV for Janus CeO<sub>2</sub> slurry.



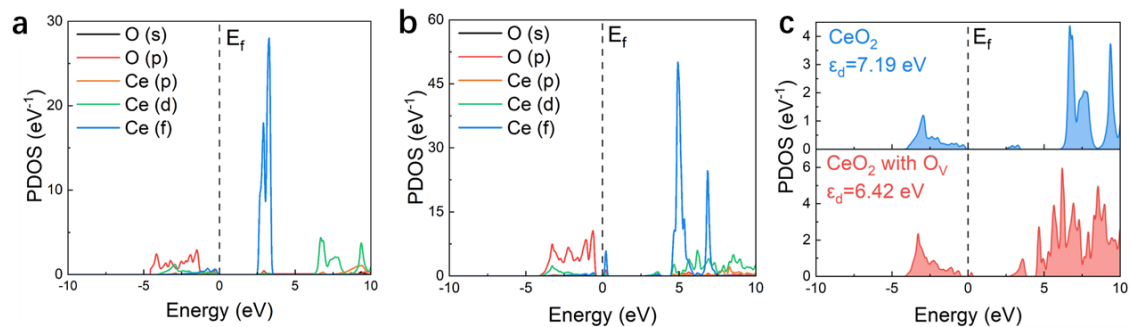
**Supplementary Fig. S26.** The band structure of CeO<sub>2</sub> particles and CeO<sub>2</sub> slurry.

**Discussion:** The constructed band structure reveals that the Janus CeO<sub>2</sub> slurry possesses a band gap of 2.67 eV, corresponding to an absorption edge of approximately 465 nm, which enables efficient excitation under 365 nm UV irradiation. Notably, the conduction band minimum (CBM) lies above the redox potential of O<sub>2</sub>/·O<sub>2</sub><sup>-</sup> (−0.33 V vs. NHE), while the valence band maximum (VBM) is below the redox potential of H<sub>2</sub>O/·OH (+2.38 V vs. NHE). This favorable band alignment facilitates the generation of both superoxide (·O<sub>2</sub><sup>-</sup>) and hydroxyl (·OH) radicals upon photoexcitation. These reactive oxygen species contribute synergistically to the oxidative enhancement observed during the PCMP process.



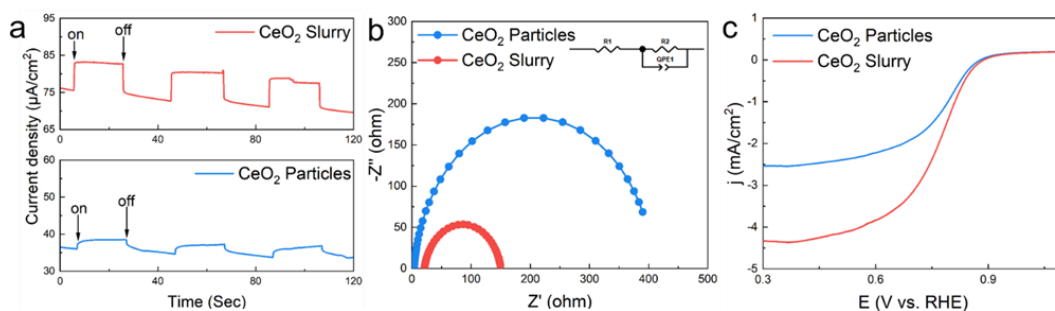
**Supplementary Fig. S27.** Time-resolved PL decay curves of CeO<sub>2</sub> particles and CeO<sub>2</sub> slurry.

**Discussion:** The transient fluorescence lifetimes of CeO<sub>2</sub> particles and Janus CeO<sub>2</sub> slurry are 4.77 ns and 5.42 ns, respectively. It indicates oxygen vacancies act as an electronic trap to induce local band structure changes in CeO<sub>2</sub>, particularly forming mid-gap states in regions with oxygen deficiency<sup>9,10</sup>. These mid-gap states act as intermediate trapping sites for electrons transitioning from the excited to the ground state, slowing the electron recombination pathway and thereby extending the lifetime of photo-generated charge carriers.



**Supplementary Fig. S28. a** Projected density of states of  $\text{CeO}_2$ . **b**  $\text{CeO}_2$  with  $\text{O}_v$ . **c** Projected density of states of Ce 3d orbitals. The Fermi level is set to zero.

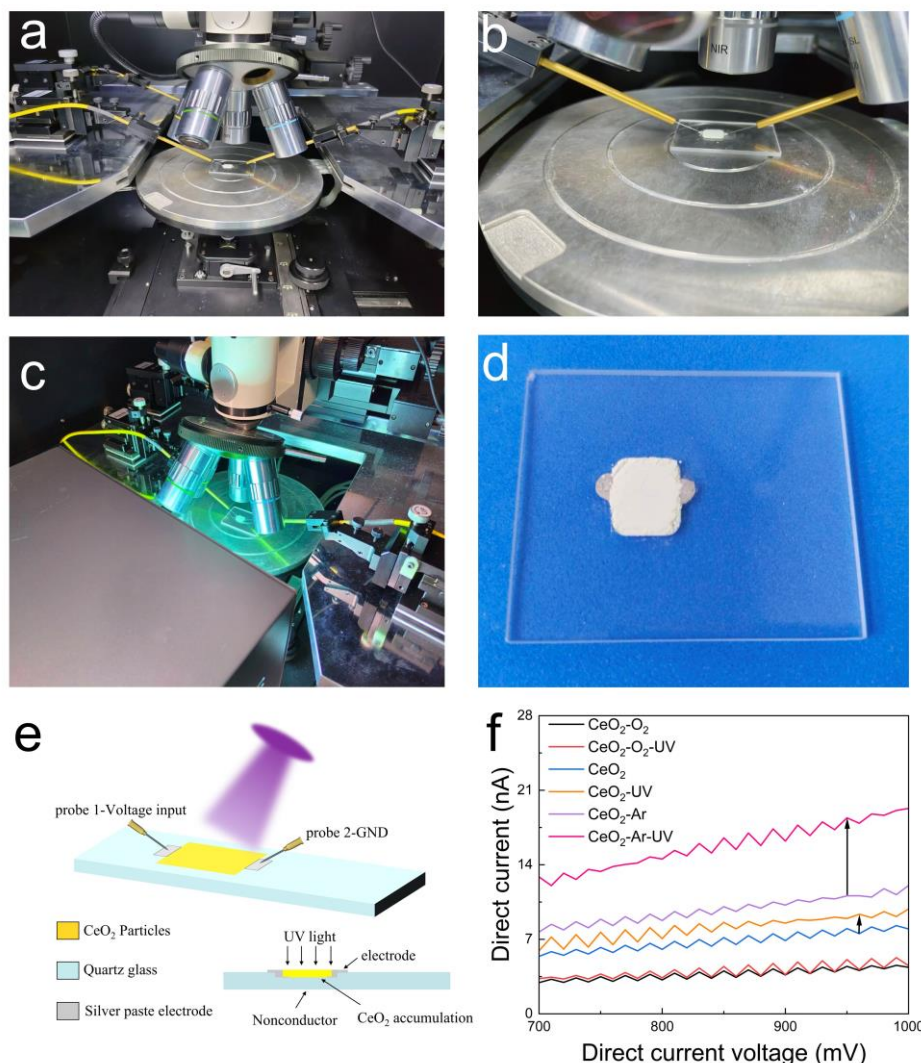
**Discussion:** The electronic structures of  $\text{CeO}_2$  with and without oxygen vacancies were analyzed by partial density of states (PDOS) calculations. It reveals that oxygen vacancies lead to excess electrons occupying the Ce 4f states, creating new electronic states, and the excess electron gain by the 4f would give rise to the charge carrier in the band. Compared to  $\text{CeO}_2$ , the d-band center of  $\text{CeO}_2$  with oxygen vacancy is closer to the 0.77 eV. Therefore, defect states cause band tailing and act as mid-states, enabling electrons to be excited from the valence band to the conduction band through multi-step excitation processes. This enhances the light utilization efficiency of  $\text{CeO}_2$  and boosts the generation rate and lifetimes of photo-generated carriers.



**Supplementary Fig. S29. a** Transient photocurrent response. **b** Polarization curves. **c** Nyquist plot of CeO<sub>2</sub> particles and CeO<sub>2</sub> slurry.

**Discussion:** The photocurrent densities over time under three cycles of intermittent illumination (Fig. S29a) demonstrated that CeO<sub>2</sub> particles exhibited weak current densities when illuminated, whereas the CeO<sub>2</sub> slurry produced a distinct photocurrent response. Upon switching off the Xe lamp, the photocurrent values of both samples rapidly declined. Notably, the photocurrent density of the CeO<sub>2</sub> slurry reached 7.55  $\mu\text{A}/\text{cm}^2$ , which was 3.07 times higher than that of CeO<sub>2</sub> particles.

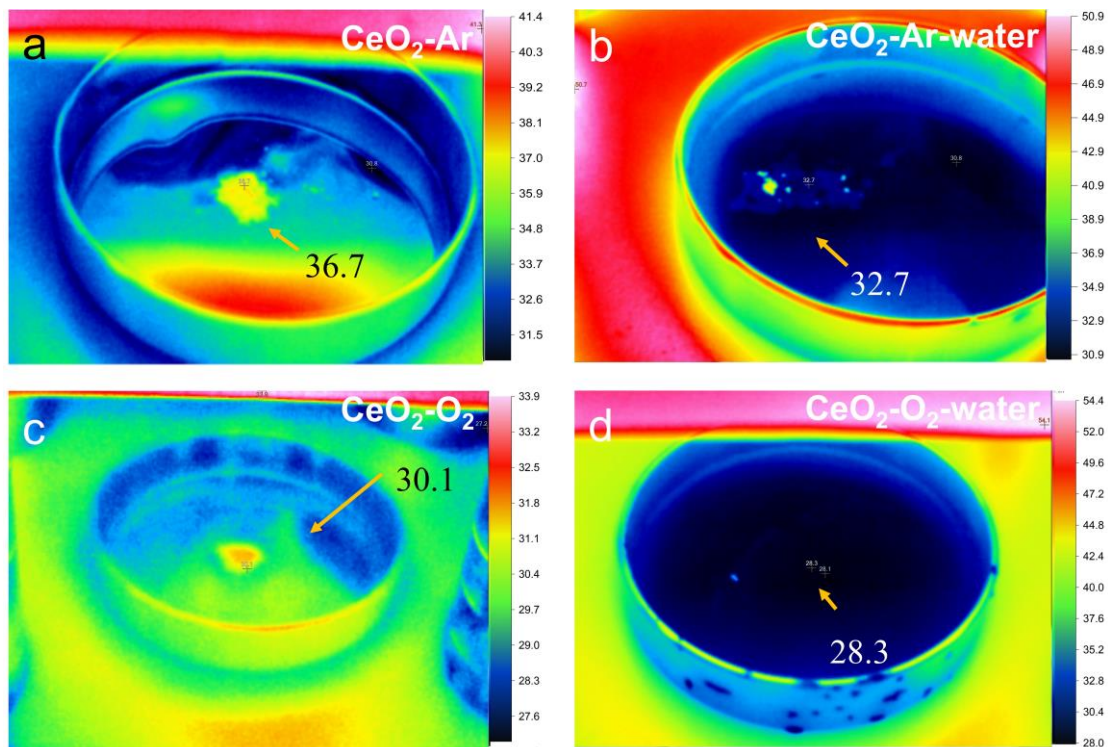
In the Nyquist plot (Fig. S29b), the semicircular radius for the CeO<sub>2</sub> slurry was smaller than that of CeO<sub>2</sub> particles, indicating lower charge transfer resistance. Additionally, the CeO<sub>2</sub> slurry displayed superior oxygen reduction reaction (ORR) performance in linear sweep voltammetry (LSV) measurements (Fig. S29c), with an onset potential of 0.87 V and a half-wave potential ( $E_{1/2}$ ) of 0.81 V, outperforming the CeO<sub>2</sub> particles (0.77 V and 0.74 V, respectively). These results highlight the highly efficient photogenerated charge transport capabilities of the CeO<sub>2</sub> slurry compared to CeO<sub>2</sub> particles.



**Supplementary Fig. S30.** **a** Schematic illustration of the I-V sweep of electrochemical electrode. **b** Probe contacting. **c** Electrochemical measurements under UV light. **d** Electrode containing cerium dioxide particles. **e** Schematic illustration of the electrochemical electrode under UV irradiation. and **f** voltage sweep from 700 mV to 1000 mV of the electrode.

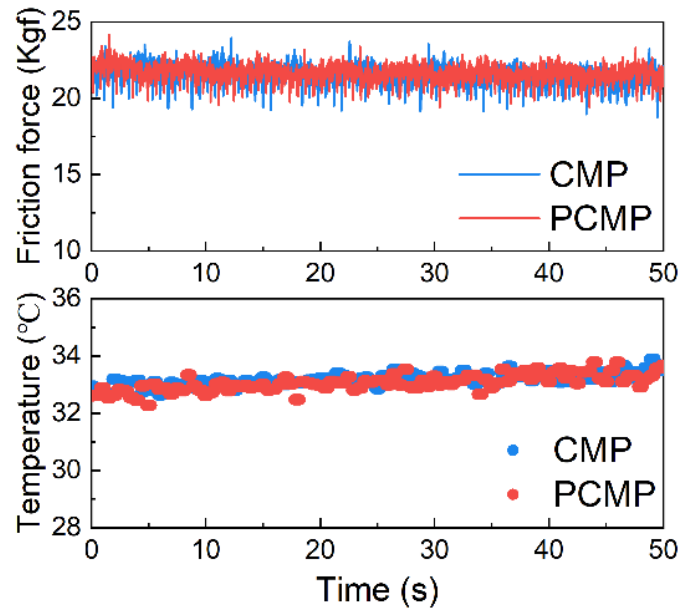
**Discussion:** The photoresponse behavior of various  $\text{CeO}_2$  were evaluated under UV irradiation using a two-probe I-V measurement setup (Fig. S30a-e). The samples were deposited on quartz glass substrates with silver paste electrodes, and UV light was applied vertically to excite photo-generated carriers. As shown in Fig. S30f, the  $\text{CeO}_2$  sample

calcined under an Ar atmosphere and subjected to UV irradiation (CeO<sub>2</sub>-Ar-UV) exhibited the highest photocurrent among all samples. This enhanced photocurrent is attributed to the high concentration of oxygen vacancies, which introduce shallow defect levels within the band gap. These defect levels act as trapping centers for photo-generated electrons, reducing carrier recombination and facilitating charge separation and transport. Additionally, the localized states enable sub-bandgap excitation and improved conductivity under UV light. In contrast, CeO<sub>2</sub> calcined in an O<sub>2</sub> atmosphere exhibited much lower photocurrent, due to the fill of oxygen vacancies, which hinders the excitation and transport of photo-generated carriers.



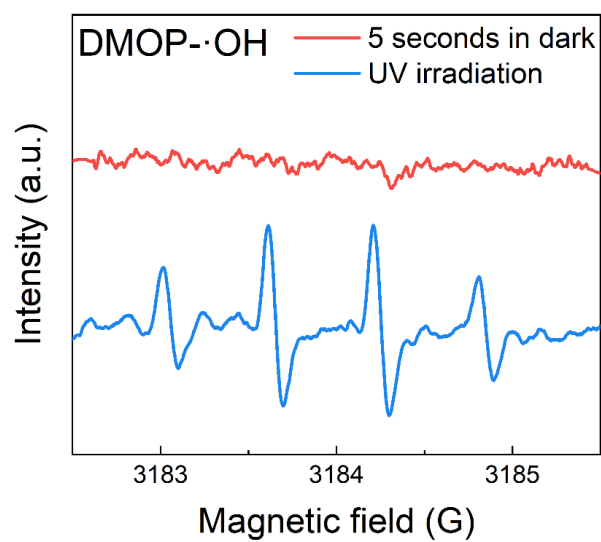
**Supplementary Fig. S31.** Infrared thermal images of CeO<sub>2</sub> samples under UV illumination. **a** CeO<sub>2</sub>-Ar, **b** CeO<sub>2</sub>-Ar-water, **c** CeO<sub>2</sub>-O<sub>2</sub>, and **d** CeO<sub>2</sub>-O<sub>2</sub>-water.

**Discussion:** The CeO<sub>2</sub>-Ar sample exhibits the highest surface temperature (36.7 °C), attributed to the presence of abundant oxygen vacancies that promote non-radiative recombination and local heating. In contrast, CeO<sub>2</sub>-O<sub>2</sub>, with fewer oxygen vacancies, shows a significantly lower temperature (30.1 °C). The addition of water reduces the surface temperature due to its cooling and shielding effects, with CeO<sub>2</sub>-O<sub>2</sub>-water reaching the lowest temperature (28.3 °C). These results highlight the role of oxygen vacancies and medium composition in regulating photothermal behavior.

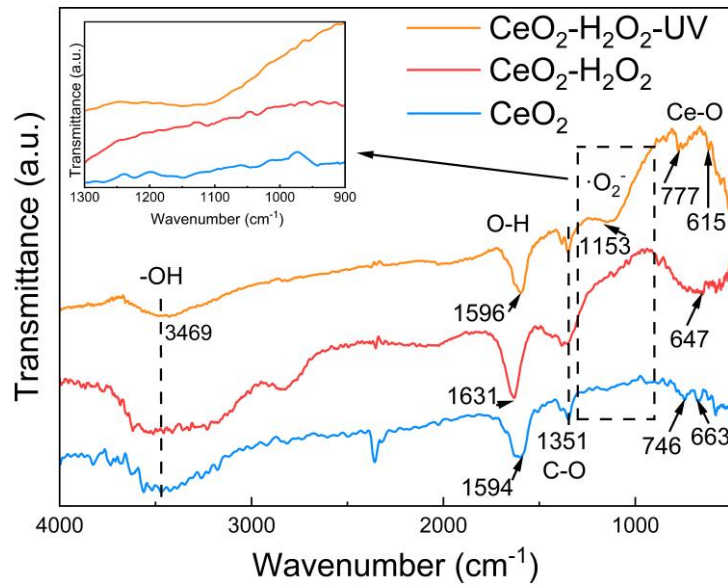


**Supplementary Fig. S32.** Friction force and slurry temperature during conventional CMP and PCMP.

**Discussion:** As shown in Fig. S32, the friction force and temperature remain relatively stable and comparable between CMP and PCMP, indicating similar polishing conditions. This is because the slurry is in dynamic flow during polishing, preventing heat accumulation in the bulk and limiting temperature rise to the  $\text{CeO}_2$  surface. The localized heating of  $\text{CeO}_2$  surface originates from non-radiative recombination of photo-generated carriers on the  $\text{CeO}_2$  surface, which releases energy as phonons and elevates the temperature at the abrasive–wafer interface.

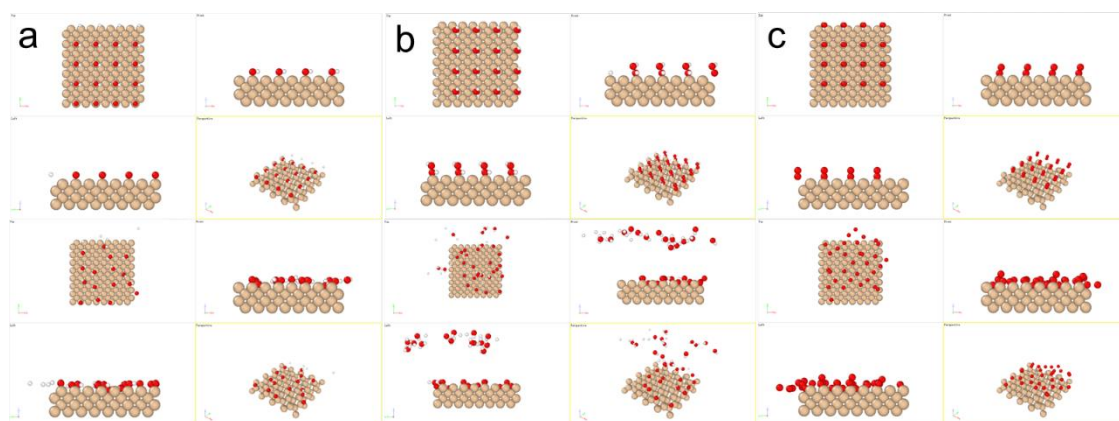


472 **Supplementary Fig. S33.** ESR spectra of DMPO-·OH for CeO<sub>2</sub> slurry under  
473 different conditions.

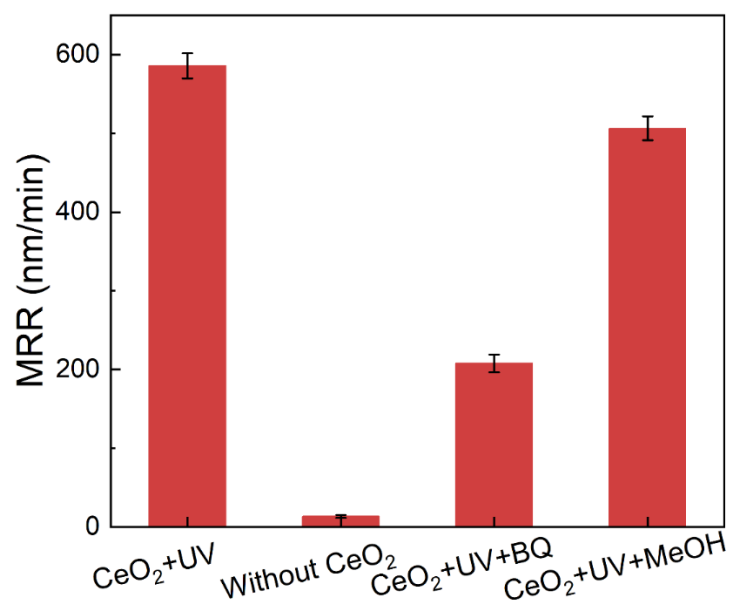


**Supplementary Fig. S34.** FTIR of CeO<sub>2</sub> under different conditions.

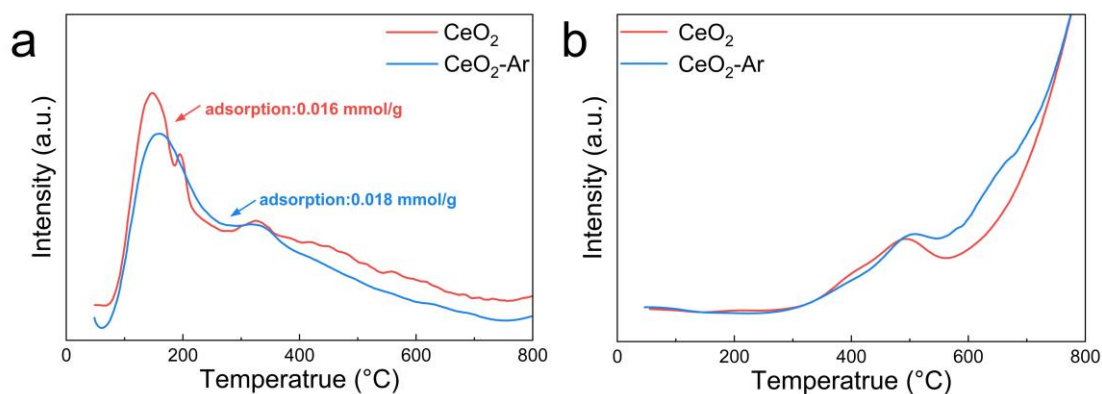
**Discussion:** A drop of H<sub>2</sub>O<sub>2</sub> was added onto the pressed CeO<sub>2</sub> film, followed by UV irradiation after a 2-minute. Compared to pristine CeO<sub>2</sub>, the CeO<sub>2</sub>-H<sub>2</sub>O<sub>2</sub> and CeO<sub>2</sub>-H<sub>2</sub>O<sub>2</sub>-UV samples exhibit several new or enhanced peaks, indicating chemical interactions between CeO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub>. The broad absorption band around 3469 cm<sup>-1</sup> is assigned to -OH stretching vibrations, while peaks near 1631 cm<sup>-1</sup> and 1596 cm<sup>-1</sup> correspond to O-H bending modes. A prominent peak at 1351 cm<sup>-1</sup> is attributed to C-O stretching. After UV irradiation, a new band appears at 1153 cm<sup>-1</sup>, which is assigned to the formation of superoxide species on the CeO<sub>2</sub> surface. The Ce-O lattice vibrations are observed in the lower wavenumber region (900–500 cm<sup>-1</sup>), with shifts from 663 cm<sup>-1</sup> and 746 cm<sup>-1</sup> to 647 cm<sup>-1</sup> and 777 cm<sup>-1</sup> in the UV-treated sample, suggesting lattice distortion and defect formation. The inset highlights the enhancement in the 900–1300 cm<sup>-1</sup> region, further confirming the generation of reactive oxygen species<sup>11</sup>.



**Supplementary Fig. S35.** Molecular dynamic simulation of silicon oxidation with (a)  $\cdot\text{OH}$ , (b)  $\text{H}_2\text{O}_2$ , and (c)  $\cdot\text{O}_2^-$ , with a molecular number of 16.



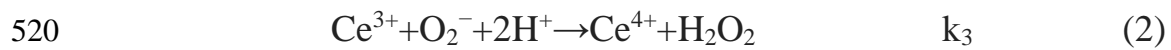
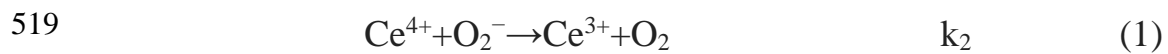
**Supplementary Fig. S36.** Comparison of MRR on Si wafers with different sacrificial agents under polishing conditions.

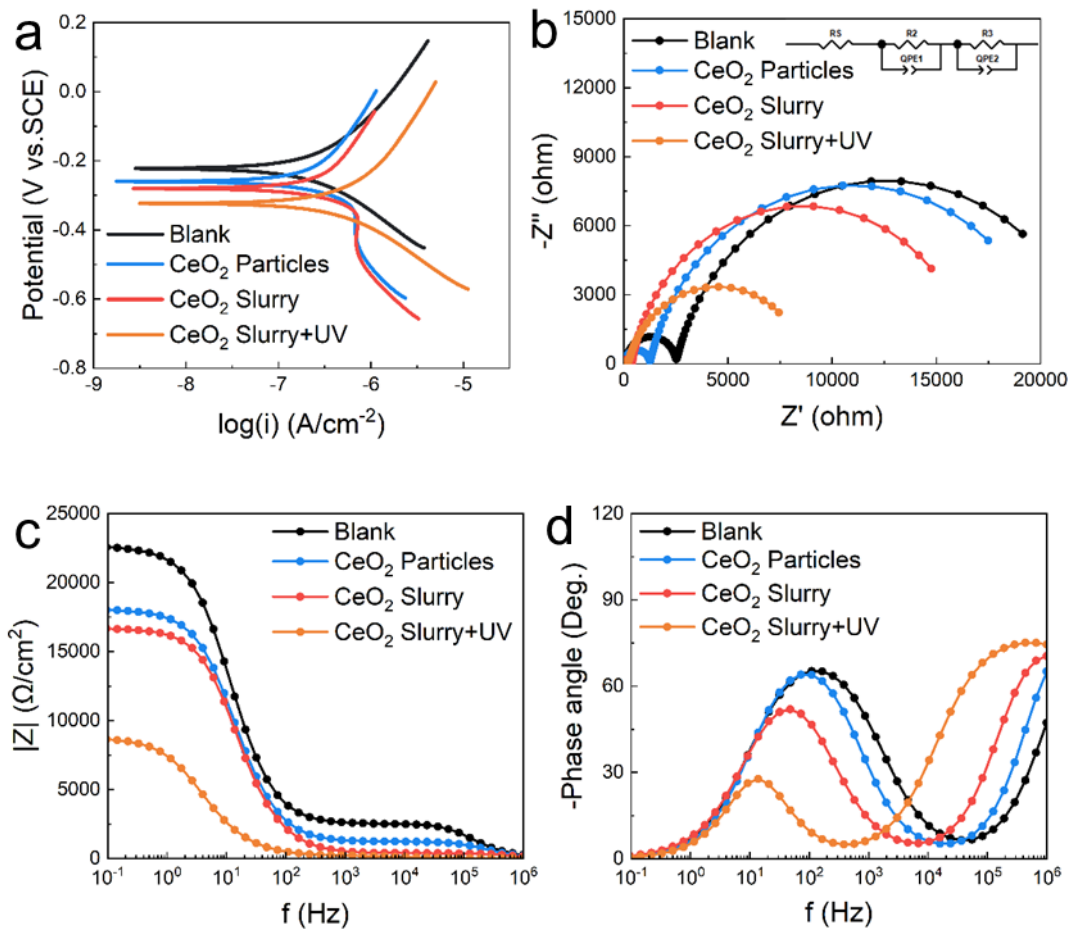


**Supplementary Fig. S37.** (a) O<sub>2</sub>-TPD profiles of CeO<sub>2</sub>, CeO<sub>2</sub>-Ar-600 showing the adsorption amounts and strength of O<sub>2</sub>. (b) H<sub>2</sub>-TPR profiles of CeO<sub>2</sub> materials showing the reducibility of reactive SOS.

**Discussion:** Since TPD and TPR measurements require pretreatment to remove surface contaminants, and the Janus CeO<sub>2</sub> slurry contains organic components such as surfactants, which can interfere with the evaluation of oxygen vacancy adsorption and desorption behavior, Ar-calcined CeO<sub>2</sub> was used for characterization. Surface oxygen species on oxide nanomaterials include surface lattice oxygen atoms as well as adsorbed reactive oxygen species such as \*O<sub>2</sub>, \*O<sub>2</sub><sup>-</sup>, and \*O<sub>2</sub><sup>2-</sup>. The influence of enhanced defect states on O<sub>2</sub> adsorption and activation was investigated using O<sub>2</sub>-temperature programmed desorption (O<sub>2</sub>-TPD), which identifies the types and strengths of oxygen adsorption. For CeO<sub>2</sub> particles, desorption of O<sub>2</sub> between 50–200 °C is attributed to weakly adsorbed \*O<sub>2</sub><sup>-</sup>, while \*O<sub>2</sub> desorbs between 150–300 °C<sup>12</sup>. As shown in Fig. S37a, compared to CeO<sub>2</sub>, oxygen-deficient CeO<sub>2</sub> exhibits weaker physical adsorption but stronger chemical adsorption between 50–250 °C, based on acidity comparisons<sup>13</sup>. Fig. S37b further demonstrates that CeO<sub>2</sub> with oxygen vacancies possesses a higher O<sub>2</sub> storage capacity, consistent with the higher values of kinetic constants  $k_2$  and  $k_3$ , facilitating the subsequent formation of superoxide radicals. Additionally, the desorption

517 curve at elevated reduction temperatures indicates that CeO<sub>2</sub>-Ar-600  
518 retains the oxidation capability associated with surface oxygen species.





**Supplementary Fig. S38.** **a** Potential polarization plots of Si electrodes in the electrolytes under different conditions. **b** Nyquist for different electrolyte conditions, with the equivalent circuit models. **c** Bode-magnitude graphs. **d** Bode-phase graphs for different conditions. Post-immersion in solutions at pH 10.

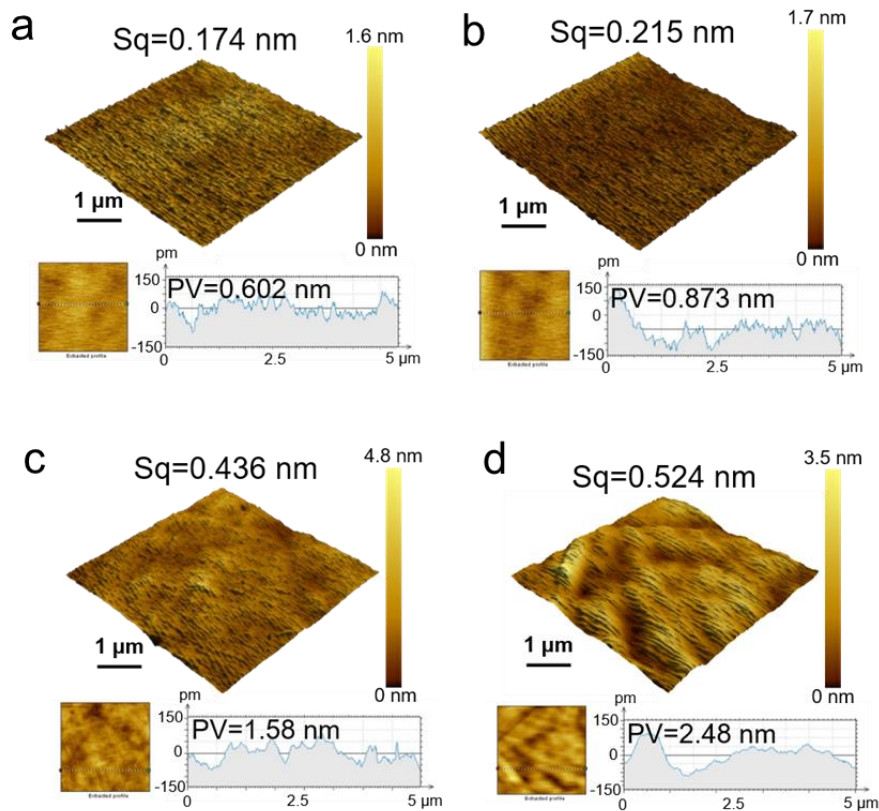
**Discussion:** To further explore the contributions of various slurry components and the influence of UV light, the oxidation processes of the Si surface were systematically analyzed using potential dynamic polarization curves (Fig. S38a). The corrosion potential ( $E_{\text{corr}}$ ) is the open-circuit potential at which the anodic (oxidation) and cathodic (reduction) reactions on a surface are balanced under steady-state conditions. A more negative  $E_{\text{corr}}$  indicates a higher tendency for the material to oxidize, meaning it is more prone to corrosion. The corrosion

current density ( $I_{\text{corr}}$ ) quantifies the rate at which corrosion proceeds. It is determined from the Tafel extrapolation of the polarization curve and is directly proportional to the corrosion rate. A higher  $I_{\text{corr}}$  means a faster corrosion rate, indicating that the material is undergoing more rapid electrochemical degradation<sup>14</sup>.

The  $E_{\text{corr}}$  and  $I_{\text{corr}}$ , derived from Tafel plots, are summarized in Table S3. In the dispersed system of  $\text{CeO}_2$  particles, the self-catalytic reaction requires significant interfacial pressure for initiation, resulting in an  $E_{\text{corr}}$  of -0.222 V and  $I_{\text{corr}}$  of  $6.561 \times 10^{-7}$  A/cm<sup>2</sup>. With the successive addition of slurry components and the introduction of UV light,  $E_{\text{corr}}$  and  $I_{\text{corr}}$  progressively increased, eventually reaching -0.323 V and  $2.067 \times 10^{-6}$  A/cm<sup>2</sup>. This progressive increase reflects the intensification of UV-induced photocatalytic reactions, which significantly accelerate the oxidation of the Si wafer.

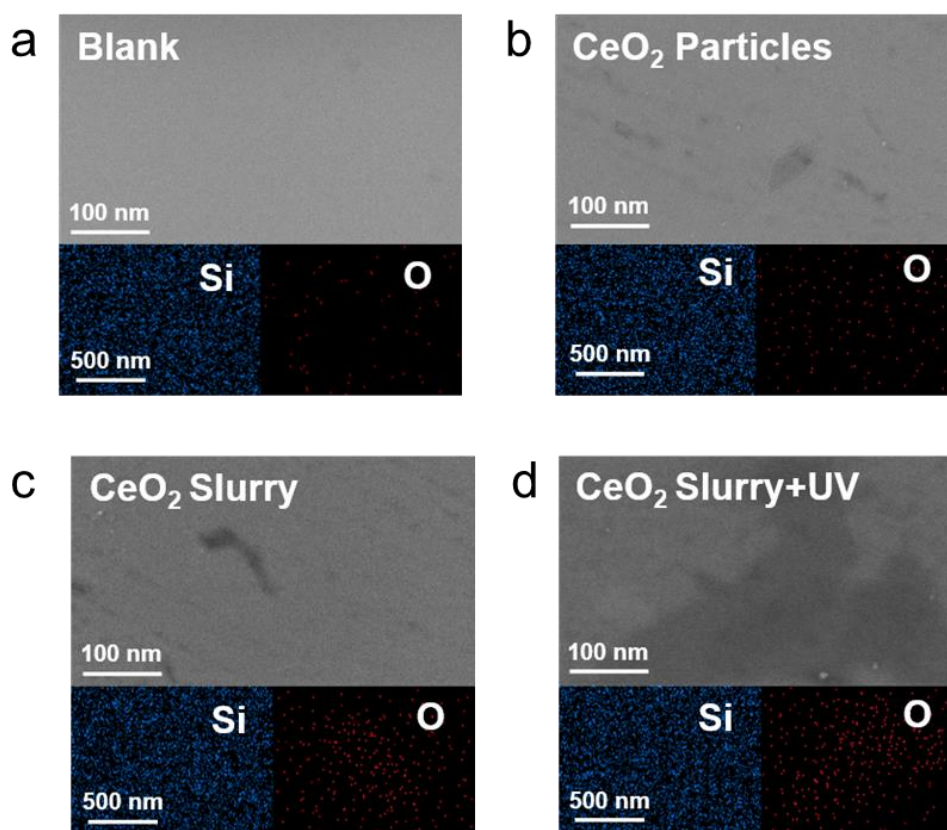
Electrochemical Impedance Spectroscopy (EIS) can probe the interfacial properties of materials by applying a small-amplitude AC voltage over a wide range of frequencies and measuring the resulting current. To further understand the interfacial dynamics, EIS parameters were analyzed using a fitted equivalent circuit model (Fig. S38b), with impedance values compiled in Table S4. The interfacial charge transfer resistance decreased through the interaction between UV light and  $\text{CeO}_2$ , reaching  $R_1 = 215 \ \Omega \cdot \text{cm}^2$ . This reduction could be attributed to generating electron-hole pairs that altered the ionic distribution at the interface between  $\text{CeO}_2$  slurry and Si wafer under UV illumination, thereby enhancing the double-layer capacitance (QPE1) to  $2.923 \times 10^{-8}$  nF/cm<sup>2</sup>. The increased capacitance decreased the low-frequency impedance, highlighting the improved interfacial charge transfer between  $\text{CeO}_2$  and Si.

562       The amplitude and phase Bode plots further corroborate this trend.  
563   Upon introducing UV light, the amplitude Bode plot (Fig. S38c)  
564   demonstrated a marked decrease in impedance modulus ( $|Z|$ ) within the  
565   low-frequency region, suggesting a reduction in charge transfer resistance  
566   ( $R_{ct}$ ). The phase Bode plot (Fig. S38d) exhibited minimal changes in the  
567   high-frequency region, predominantly influenced by the double-layer  
568   capacitance ( $C_{dl}$ ). However, the phase angle decreased noticeably in the  
569   mid-to-low frequency range, indicating a transition from capacitive  
570   behavior to enhanced oxidation activity for the Si wafer.



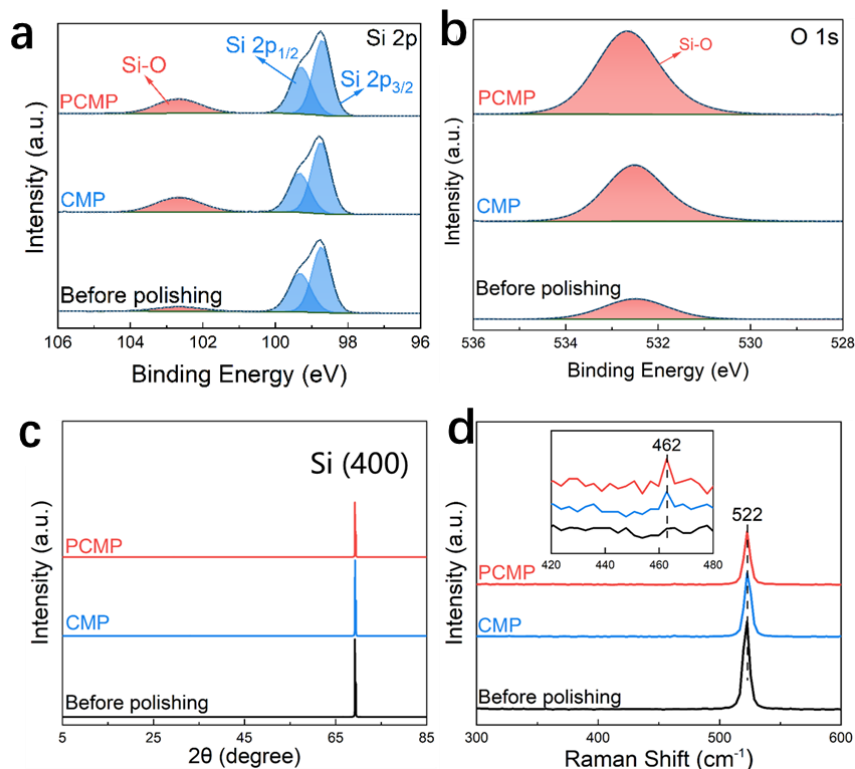
**Supplementary Fig. S39.** The 3D AFM images with corresponding height profiles for **a** Si wafer before immersion, **b** CeO<sub>2</sub> particles, **c** CeO<sub>2</sub> slurry, and **d** CeO<sub>2</sub> slurry with UV illumination.

**Discussion:** AFM investigations visually supported the oxidation processes. The Sq of the initial Si wafer was measured at 0.174 nm, with a PV of 0.602 nm, indicating a relatively smooth Si surface with minimal evidence of corrosion or surface alteration (Fig. S39a). With the progressive introduction of CeO<sub>2</sub> particles, slurry components, and UV light, the Sq increased gradually, reaching a final value of 0.524 nm, and the peak-to-valley height reached 2.48 nm (Fig. S39b-d).



**Supplementary Fig. S40.** The SEM images, and EDS elemental mapping for Si and O are shown for **a** Si wafer before immersion, **b** CeO<sub>2</sub> particles, **c** CeO<sub>2</sub> slurry, and **d** CeO<sub>2</sub> slurry with UV illumination.

**Discussion:** SEM analyses further confirmed the formation of an oxidation layer (Fig. S40), with elemental mapping showing a significant increase in oxygen content (Table S5). Compared to the blank sample, the oxygen content (both in weight and atomic percentages) increases progressively with the addition of CeO<sub>2</sub> particles, CeO<sub>2</sub> slurry, and CeO<sub>2</sub> slurry under UV irradiation. Specifically, the atomic percentage of oxygen increases from 3.31% (blank) to 14.69% (CeO<sub>2</sub> slurry + UV), indicating a substantial enhancement in surface oxidation.

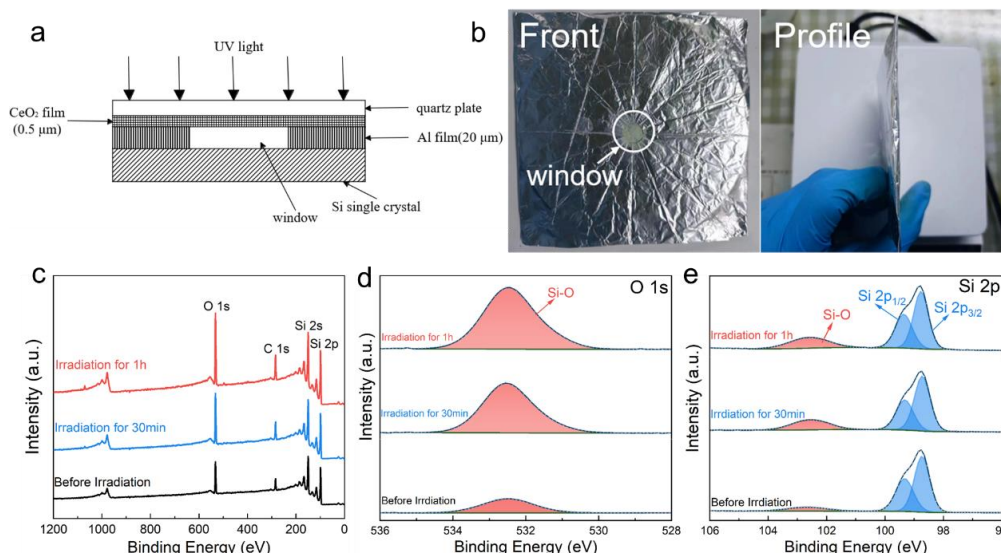


Supplementary Fig. S41. **a** Si 2p, **b** O 1s XPS spectra of Si wafers surface under different conditions. **c** XRD spectra. **d** Raman spectra of Si wafers surface under different conditions.

**Discussion:** XPS was conducted on Si wafers after CMP, and after PCMP to evaluate the surface oxygen content. In the Si 2p spectra (Fig. S41a), peaks at 98.70 eV and 99.30 eV were attributed to monocrystalline Si. In contrast, peaks at 102.58 eV in the Si 2p and 531.78 eV in the O 1s spectra (Fig. S41b) were attributed to Si-O bonds, confirming the oxidation processes during CMP and PCMP.

XRD patterns of the Si surface before polishing, after CMP, and after PCMP show a single diffraction peak at 69.63° corresponding to the Si (400) plane, with no additional peaks observed for SiO<sub>2</sub>. This absence is attributed to the extremely thin and poorly crystalline nature of the surface oxide layer, making it undetectable by XRD (Fig. S41c). In contrast, Raman spectroscopy reveals a clear signal at 462 cm<sup>-1</sup> attributed

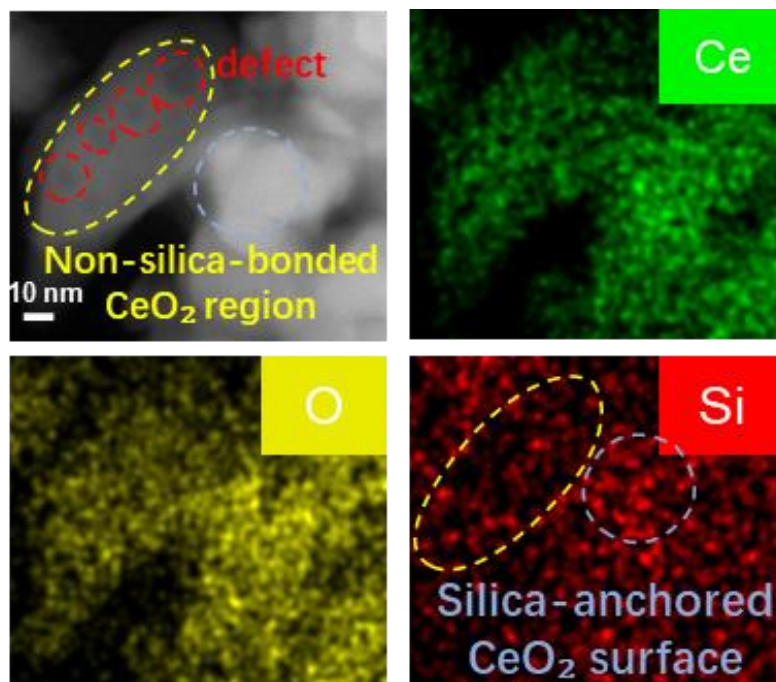
610 to the symmetric stretching vibration of the Si-O-Si bond, corresponding  
611 to the vibrational mode of amorphous silicon oxide (Fig. S41d). This  
612 peak is especially evident after PCMP treatment, indicating the formation  
613 of an oxide layer. The intensity ratio of the SiO<sub>2</sub> layer to monocrystalline  
614 silicon after CMP is 0.0237. In contrast, this ratio increases to 0.0363  
615 after photocatalytic CMP, suggesting increased SiO<sub>2</sub> layer thickness.



**Supplementary Fig. S42.** **a** Schematic illustration of the noncontact photocatalytic oxidation system for the Si wafer surface. **b** Digital image of the noncontact photocatalytic oxidation. **c** XPS spectra, **d** O 1s XPS spectra. **e** Si 2p XPS spectra of Si wafer surface under different radiation times.

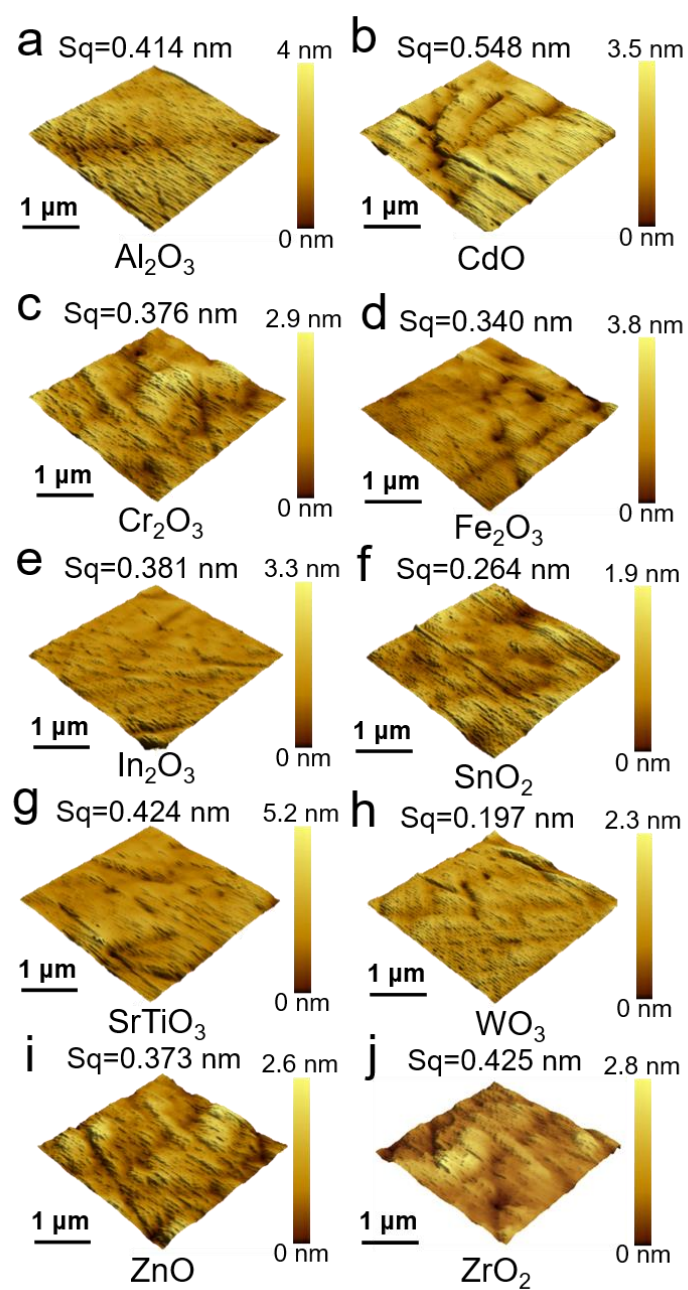
**Discussion:** An aluminum metal film, approximately 20 μm thick, was placed between the quartz plate coated with the CeO<sub>2</sub> film and the Si substrate to maintain a gap distance of 20 μm between the CeO<sub>2</sub> film and the Si surface, ensuring noncontact conditions<sup>15</sup>.

XPS spectra of the Si surface after irradiation for 0 h, 0.5 h, and 1 h demonstrated that SiO<sub>2</sub> formation could occur on the Si surface via photocatalytic oxidation even without direct contact between the CeO<sub>2</sub> film and the Si surface. The peaks at 532.40 and 532.20 eV binding energies in the O 1s XPS spectrum correspond to SiO<sub>2</sub> and Si-O, respectively. In the Si 2p XPS spectrum, the peaks at 98.70 and 99.30 eV are attributed to Si, while the peaks at 103.30 and 102.40 eV correspond to SiO<sub>2</sub> and Si-O, respectively. These results indicate that, even under noncontact conditions, the Si surface undergoes oxidation to form SiO<sub>2</sub> via photocatalytic activity from CeO<sub>2</sub>, with longer irradiation times resulting in a more pronounced oxidation effect.

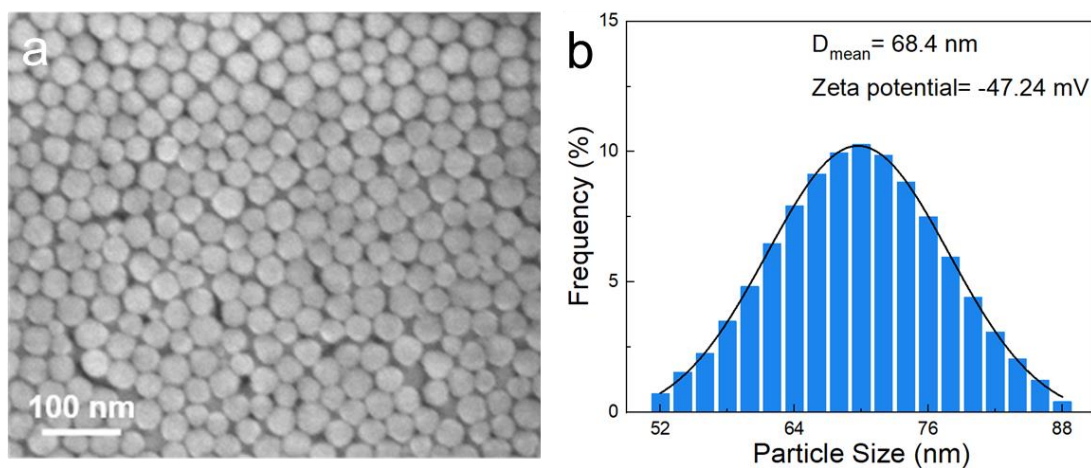


**Supplementary Fig. S43.** HAADF-STEM image and corresponding EDS elemental mappings of Ce (green), O (yellow), and Si (red) for CeO<sub>2</sub> particles after polishing a silicon wafer.

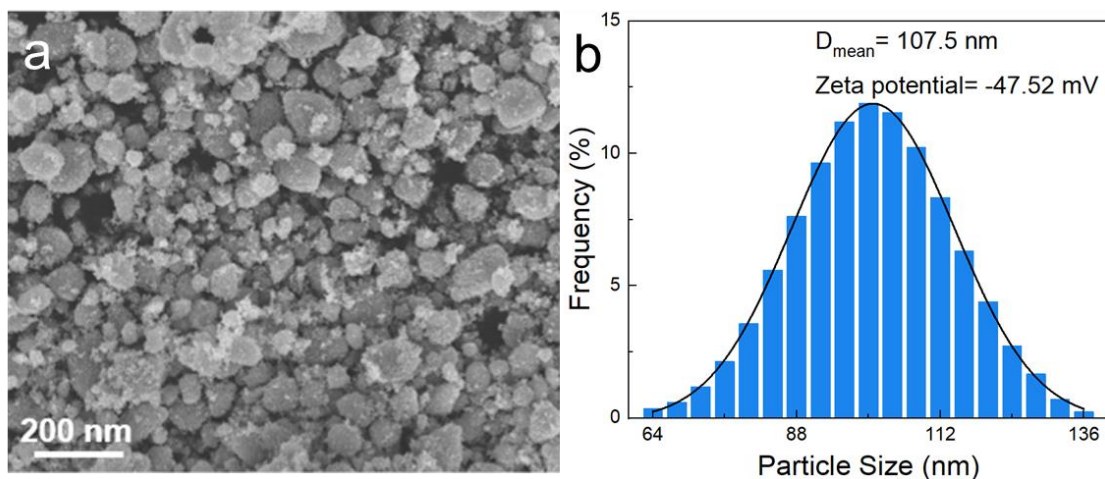
**Discussion:** To elucidate the interfacial interactions between CeO<sub>2</sub> abrasives and the silicon wafer during PCMP, HAADF-STEM and EDS mapping were performed on CeO<sub>2</sub> after polishing. The yellow dashed region marks a CeO<sub>2</sub> domain not bonded to silica, characterized by apparent structural defects. In contrast, the blue dashed region shows a silica-anchored CeO<sub>2</sub> surface, where strong co-localization of Ce, O, and Si signals suggests the formation of interfacial Ce–O–Si bonding.



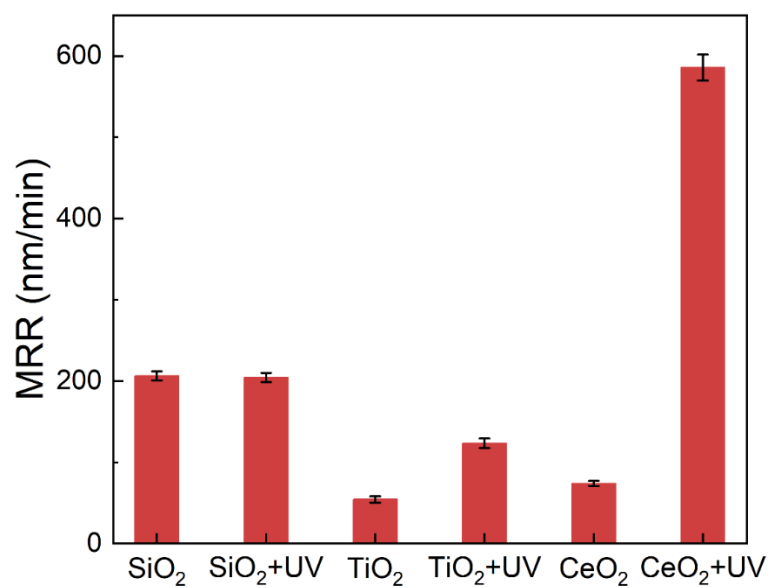
647 **Supplementary Fig. S44.** 3D AFM images of Si wafer surfaces polished under  
 648 different abrasives: **a**  $\text{Al}_2\text{O}_3$ ; **b** CdO; **c**  $\text{Cr}_2\text{O}_3$ ; **d**  $\text{Fe}_2\text{O}_3$ ; **e**  $\text{In}_2\text{O}_3$ ; **f**  $\text{SnO}_2$ ; **g**  $\text{SrTiO}_3$ ; **h**  
 649  $\text{WO}_3$ ; **i** ZnO; **j**  $\text{ZrO}_2$ .



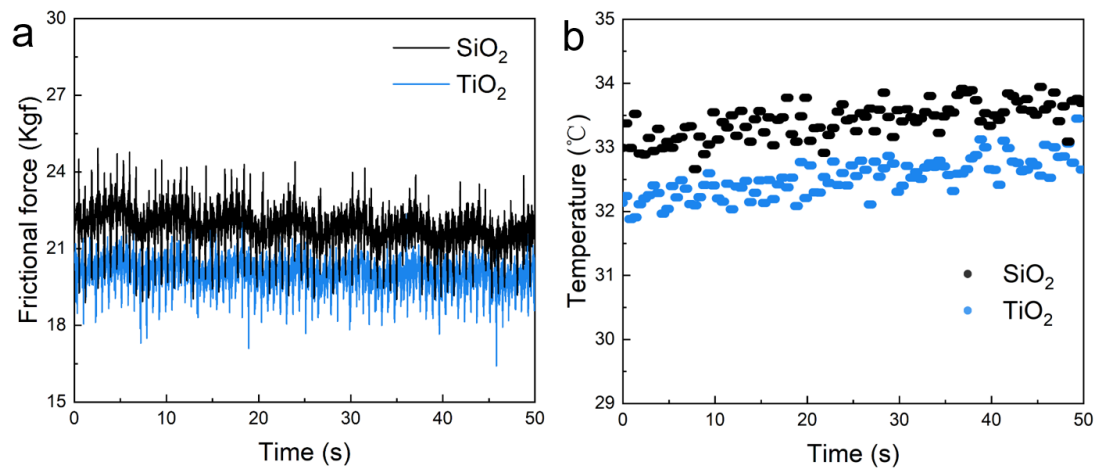
650 **Supplementary Fig. S45. a** SEM image of nano SiO<sub>2</sub>. **b** Particle size distribution  
 651 histogram and zeta potential of 2.5 wt.% SiO<sub>2</sub> exhibits an average SiO<sub>2</sub> particle size of  
 652 68.4 nm and a zeta potential of -47.24 mV.



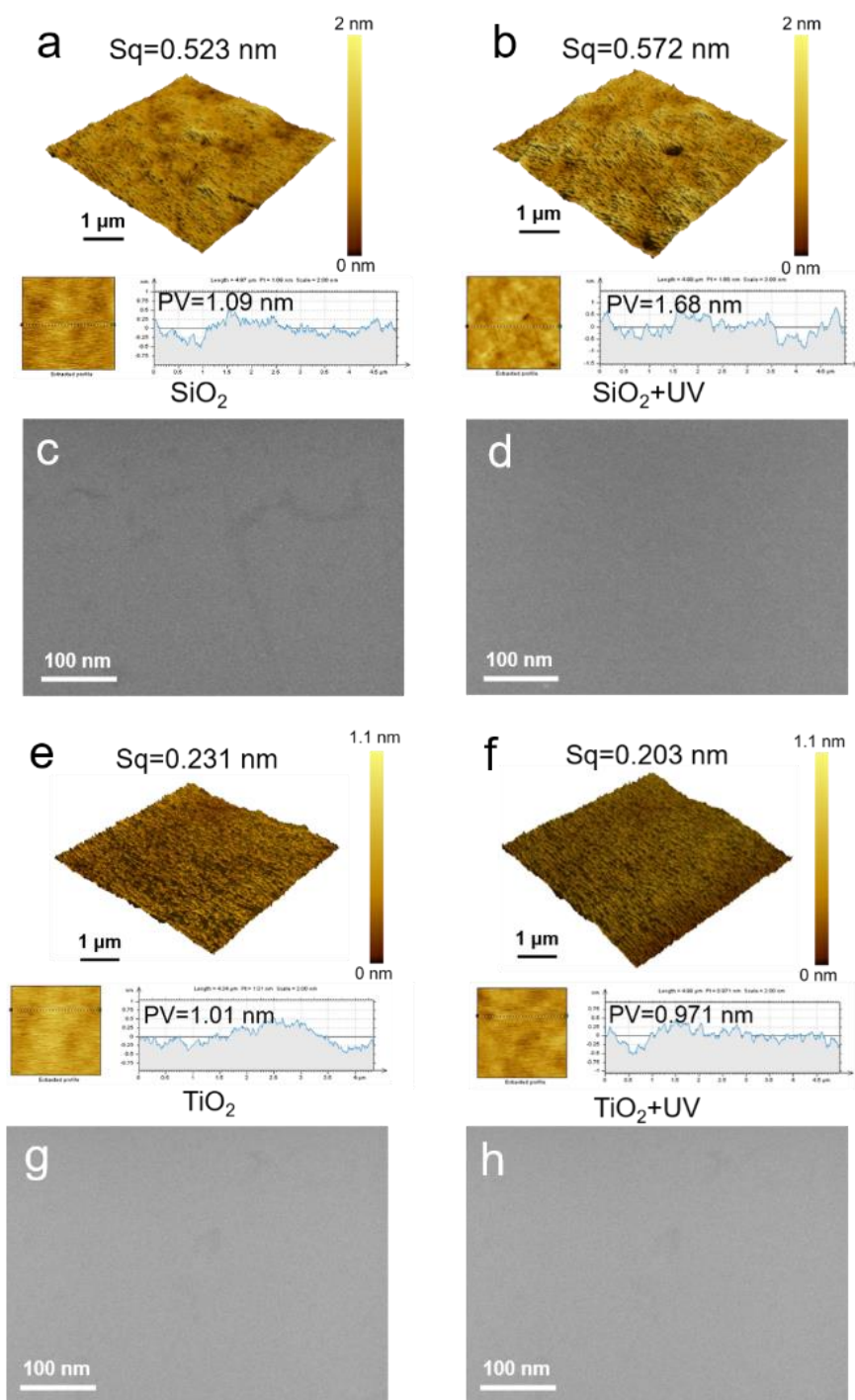
653 **Supplementary Fig. S46. a** SEM image of nano  $\text{TiO}_2$ . **b** Particle size distribution  
 654 histogram and zeta potential of 2.5 wt.%  $\text{TiO}_2$  exhibits an average  $\text{TiO}_2$  particle size  
 655 of 107.5 nm and a zeta potential of -47.52 mV.



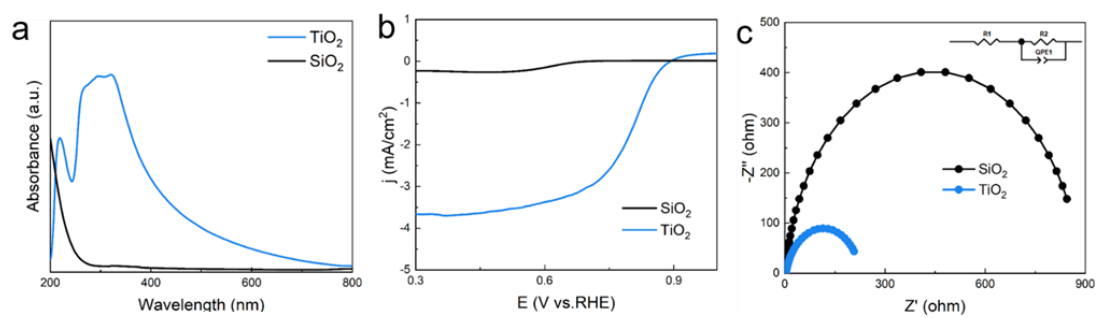
656 **Supplementary Fig. S47.** Comparison of MRR on the Si wafers by different slurries  
657 (SiO<sub>2</sub>, TiO<sub>2</sub>, CeO<sub>2</sub>) containing 2.5 wt% H<sub>2</sub>O<sub>2</sub> under various conditions of UV light at  
658 pH 10.



659 **Supplementary Fig. S48. a** Surface friction force. **b** Temperature of abrasives in  
 660 polishing Si wafers.



661 **Supplementary Fig. S49.** SEM images, 3D AFM images, and height profiles of Si  
 662 wafer surfaces polished under different conditions: **a, c** 2.5 wt% SiO<sub>2</sub>; **b, d** 2.5 wt%  
 663 SiO<sub>2</sub>; **e, g** 2.5 wt% TiO<sub>2</sub>; **f, h** 2.5 wt% TiO<sub>2</sub> with UV light irradiation.



664 **Supplementary Fig. S50. a** UV-Vis spectra. **b** Polarization curves. **c** Nyquist plot of  
 665  $\text{SiO}_2$  and  $\text{TiO}_2$ .

**Supplementary Tables**

**Supplementary Table S1.** Percentage of different substances obtained by Ce 3d and O 1s spectroscopy.

| Sample                     | Ce <sup>3+</sup> (%) | Ce <sup>4+</sup> (%) | Ov (%) |
|----------------------------|----------------------|----------------------|--------|
| CeO <sub>2</sub> particles | 21.34                | 78.66                | 30.17  |
| CeO <sub>2</sub> slurry    | 26.52                | 73.48                | 34.02  |

671 **Supplementary Table S2.** EXAFS fitting parameters at the Ce L<sub>3</sub>-edge for various  
672 samples.

| Sample                        | Shell | $CN^a$ | $R(\text{\AA})^b$ | $\sigma(\text{\AA}^2)^c$ | $\Delta E_0(\text{eV})^d$ | $R$<br>factor |
|-------------------------------|-------|--------|-------------------|--------------------------|---------------------------|---------------|
| CeO <sub>2</sub><br>reference | Ce-O  | 8      | 2.31              | 0.015                    | 5.26                      | 0.015         |
| CeO <sub>2</sub><br>particles | Ce-O  | 4.97   | 2.32              | 0.0091                   | 6.32                      | 0.019         |
| CeO <sub>2</sub> slurry       | Ce-O  | 4.40   | 2.34              | 0.0103                   | 6.82                      | 0.014         |

673 <sup>a</sup> $CN$ , coordination number; <sup>b</sup> $R(\text{\AA})$ , the distance to the neighboring atom; <sup>c</sup> $\sigma(\text{\AA}^2)$ ,  
674 Debye-Waller factor to account for both thermal and structural disorders; <sup>d</sup> $\Delta E_0$  inner  
675 potential correction;  $R$  factor indicates the goodness of the fit.

676 **Supplementary Table S3.**  $E_{\text{corr}}$  and  $I_{\text{corr}}$  of silicon in different compositions of slurry.

| Component                  | $E_{\text{corr}}$ vs. SCE (V) | $I_{\text{corr}}$ (A/cm <sup>2</sup> ) |
|----------------------------|-------------------------------|--|
| Blank                      | -0.222                        | $6.561 \times 10^{-7}$                 |
| CeO <sub>2</sub> particles | -0.257                        | $1.143 \times 10^{-6}$                 |
| CeO <sub>2</sub> slurry    | -0.278                        | $2.237 \times 10^{-6}$                 |
| CeO <sub>2</sub> slurry+UV | -0.323                        | $4.502 \times 10^{-6}$                 |

677

678 **Supplementary Table S4.** Impedance parameters of silicon in slurries with different  
679 compositions obtained by EEC fitting.

| Component                     | <b>Rs</b><br>( $\Omega \cdot \text{cm}^2$ ) | <b>R1</b><br>( $\Omega \cdot \text{cm}^2$ ) | <b>R2</b><br>( $\Omega \cdot \text{cm}^2$ ) | <b>QPE1</b><br>(nF/cm <sup>2</sup> ) | <b>QPE2</b><br>(nF/cm <sup>2</sup> ) |
|-------------------------------|---|---|---|--------------------------------------|--------------------------------------|
| Blank                         | 102.32                                      | 2398  | 20080                                       | $8.793 \times 10^{-10}$              | 0.981                                |
| CeO <sub>2</sub><br>particles | 29.21                                       | 1231  | 19316                                       | $1.241 \times 10^{-9}$               | 0.974                                |
| CeO <sub>2</sub> slurry       | 6.99  | 394   | 16307                                       | $2.261 \times 10^{-9}$               | 0.951                                |
| CeO <sub>2</sub><br>slurry+UV | 6.61  | 215   | 8253  | $2.923 \times 10^{-8}$               | 0.927                                |

680

681 **Supplementary Table S5.** EDS of silicon in different compositions of slurry.

| Component                     | wt%   |      | Atomic percentage |       |
|-------------------------------|-------|------|-------------------|-------|
|                               | Si    | O    | Si                | O     |
| Blank                         | 98.37 | 1.63 | 96.69             | 3.31  |
| CeO <sub>2</sub> particles    | 97.65 | 2.35 | 95.88             | 4.12  |
| CeO <sub>2</sub> slurry       | 94.22 | 5.78 | 90.35             | 9.65  |
| CeO <sub>2</sub><br>slurry+UV | 91.01 | 8.99 | 85.31             | 14.69 |

682

683 **Supplementary Table S6.** Energy-related parameters are calculated through  
 684 molecular dynamics simulation.

|                               | $\Delta H/\text{eV}$<br>(Total) | $\Delta H/\text{eV}$ (Per<br>group) | $T \cdot \Delta S/\text{eV}$ (Per<br>group) | $\Delta G/\text{eV}$ (Per<br>group) |
|-------------------------------|---------------------------------|-------------------------------------|---|-------------------------------------|
| H <sub>2</sub> O <sub>2</sub> | -75.34                          | -4.70                               | -0.53                                       | -4.17                               |
| ·OH                           | -26.76                          | -1.67                               | -0.72                                       | -0.94                               |
| ·O <sub>2</sub> <sup>-</sup>  | -161.69                         | -10.10                              | -0.62                                       | -9.48                               |

685

686 **Supplementary Table S7.** Performance comparison under different polishing  
687 conditions for Si wafers.

| Slurry component  | Polishing conditions  | MRR<br>(nm/min) | Surface<br>roughness<br>(nm) | Ret. |
|---|---|-----------------|------------------------------|------|
| CeO <sub>2</sub> /Na <sub>4</sub> P <sub>2</sub> O <sub>7</sub> /H <sub>2</sub> O <sub>2</sub>                                | 90 rpm,70 rpm,30 Kpa,<br>15 mL/min,pH=10                      | 127             | 0.067                        | 16   |
| SiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub>   | 40 rpm,40 rpm,7 psi,<br>7 L/min,alkaline                      | 945             | 0.94                         | 17   |
| SiO <sub>2</sub> /KNO <sub>3</sub> /H <sub>2</sub> O <sub>2</sub>   | 87 rpm,80 rpm,2.8 psi,<br>80 mL/min,pH=10.5                   | 177.87          | 0.887                        | 18   |
| SiO <sub>2</sub> /NH <sup>4+</sup> /H <sub>2</sub> O <sub>2</sub>   | 87 rpm,80 rpm,2.5 psi,<br>80 mL/min,pH=10.5                   | 168.7           | 0.744                        | 19   |
| amino-modified SiO <sub>2</sub>   | 80 rpm,80 rpm,2.64 psi,<br>90 g/min,pH=10.6                   | 132.53          | 0.219                        | 20   |
| SiO <sub>2</sub> /EDA/H <sub>2</sub> O <sub>2</sub>   | 71 rpm,69 rpm,5.7 psi,<br>100 mL/min,pH=10.9                  | 552.8           | 0.353                        | 21   |
| SiO <sub>2</sub> /C <sub>6</sub> H <sub>5</sub> O <sub>7</sub> (NH <sub>4</sub> ) <sub>3</sub> /H <sub>2</sub> O <sub>2</sub> | 87 rpm,80 rpm,2.8 psi,<br>80 mL/min,pH=10.5                   | 197.98          | 0.563                        | 22   |
| CeO <sub>2</sub> @ZIF-8/PVP   | 87 rpm,80 rpm,80 N,<br>100 mL/min                             | 151.07          | 0.231                        | 23   |
| SiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub>   | 180 rpm,16.7 KPa,10<br>mL/min,pH=9                            | 60              | 0.4                          | 24   |
| CeO <sub>2</sub> /PhPs  | 180 rpm,16.7 KPa,10<br>mL/min,pH=9, UV<br>irradiation(365 nm) | 23              | 0.19                         | 24   |
| SiO <sub>2</sub> /MEA/H <sub>2</sub> O <sub>2</sub>   | 40 rpm,40 rpm,2 psi,155<br>mL/min,pH=10.5                     | 24              | 0.178                        | 25   |

|  |   |        |        |              |
|--|---|--------|--------|--------------|
| CeO <sub>2</sub> @GO/PVP                             | 87 rpm,80 rpm,80<br>N,100 mL/min,pH=7-8                               | 190.58 | 0.278  | 26           |
| Nd-doped CeO <sub>2</sub> /PVP                       | 120 rpm,90 rpm,10 N,<br>100 mL/min,pH=6-6.5                           | 138.05 | 0.468  | 27           |
| SiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub>      | 30 rpm,30 rpm,4 psi,210<br>mL/min,pH=11                               | 265    | 0.175  | 28           |
| SiO <sub>2</sub> /TMAH/CPAM/<br>AEO-9/PVP            | 60 rpm,60 rpm,1.5 psi,<br>300 mL/min                                  | 13     | 0.447  | 29           |
| SiO <sub>2</sub> /MEA/H <sub>2</sub> O <sub>2</sub>  | 93 rpm,87 rpm,2 psi,250<br>mL/min,pH=11.5                             | 680    | 0.299  | 30           |
| SiO <sub>2</sub> /MEA/H <sub>2</sub> O <sub>2</sub>  | 93 rpm,87 rpm,2 psi,250<br>mL/min,pH=11.5,                            | 650    | 0.225  | 30           |
| CeO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub> /PVP | 93 rpm,87 rpm,4.75 psi,<br>30 mL/min,pH=10, UV<br>irradiation(365 nm) | 585.8  | 0.0437 | This<br>work |

689 **Supplementary Table S8.** The average friction force and temperature of Si wafers  
690 polished with SiO<sub>2</sub>, CeO<sub>2</sub>, TiO<sub>2</sub> and CeO<sub>2</sub> slurry.

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| Abrasive                   | Friction force (Kgf) | Temperature (°C) |
|----------------------------|----------------------|------------------|
| <hr/>                      |                      |                  |
| SiO <sub>2</sub>           | 23.33                | 32.62            |
| CeO <sub>2</sub> particles | 21.65                | 33.08            |
| TiO <sub>2</sub>           | 20.08                | 33.49            |
| CeO <sub>2</sub> slurry    | 21.63                | 32.97            |

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691

**Supplementary Table S9.** Impedance parameters of different abrasives obtained by EEC fitting.

| Abrasive                   | R1 ( $\Omega \cdot \text{cm}^2$ ) | R2 ( $\Omega \cdot \text{cm}^2$ ) | QPE1 (nF/cm <sup>2</sup> ) |
|----------------------------|-----------------------------------|-----------------------------------|----------------------------|
| SiO <sub>2</sub>           | 3.47                              | 869.7                             | $4.0502 \times 10^{-5}$    |
| CeO <sub>2</sub> particles | 3.76                              | 398.2                             | $3.0421 \times 10^{-5}$    |
| TiO <sub>2</sub>           | 4.65                              | 220.6                             | $2.2264 \times 10^{-5}$    |
| CeO <sub>2</sub> slurry    | 21.76                             | 121.3                             | $1.5176 \times 10^{-5}$    |

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