

Supporting Information

Photochemical-Co-Mechanical Polishing for Silicon Wafer Using a Janus CeO₂ Slurry

5
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16 **Supplementary Methods**

17 **Materials**

18 All solutions used in this study were prepared with deionized water,
19 and the abrasive was cerium oxide (99.95% purity, supplied by Shanghai
20 Aladdin Biochemical Technology Co., Ltd.). All chemical reagents were
21 of analytical grade. Unless otherwise specified, all solutions in this paper
22 have a pH of 10. Chemical agents utilized encompass: hydrogen peroxide
23 (H_2O_2 ; with a purity of 30%, supplied by Tianjin Fengchuan Chemical
24 Reagent Technology Co., Ltd.), PVP 99% pure, supplied by Shanghai
25 Aladdin Biochemical Technology Co., Ltd.), silica sol (40% purity,
26 supplied by Jinwei Group Co., Ltd.). Titanium Oxide (TiO_2 ; 99.9% pure,
27 supplied by Shanghai Aladdin Biochemical Technology Co., Ltd.),
28 aluminium oxide (Al_2O_3 ; 99% pure, supplied by Shanghai Dibo
29 Chemicals Technology Co., Ltd.), ferric oxide (Fe_2O_3 ; 99.5% pure,
30 supplied by Shanghai Dibo Chemicals Technology Co., Ltd.), zinc oxide
31 (ZnO ; 99.9% pure, supplied by Meryer (Shanghai) Biochemical
32 Technology Co., Ltd.), strontium titanate (SrTiO_3 ; 99% pure, supplied by
33 Bide Pharmatech Co., Ltd.), tungsten trioxide (WO_3 ; 99.8% pure,
34 supplied by Anhui Zesheng Technology Co., Ltd.), stannic oxide (SnO_2 ;
35 99.5% pure, supplied by Shanghai Macklin Biochemical Technology Co.,
36 Ltd.), cadmium oxide (CdO ; 99.99% pure, supplied by Beijing innoChem
37 Science & Technology Co., Ltd.), zirconium dioxide (ZrO_2 ; 99.99% pure,
38 supplied by Mreda Technology Co., Ltd.), benzoquinone (BQ, $\text{C}_6\text{H}_4\text{O}_2$;
39 99% pure, supplied by Mreda Technology Co., Ltd.), methanol (MeOH,
40 CH_4O ; 99% pure, supplied by Shanghai Macklin Biochemical
41 Technology Co., Ltd.), perfluorosulfonic acid-PTFE copolymer (nafion;
42 5% pure, supplied by E.I.Du Pont Company), potassium hydroxide

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

45

46 **Electrochemical Experiment**

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

50 The corrosion properties of Si in designated solutions were investigated.
51 Data for open circuit potential (OCP), dynamic potential polarization
52 (Tafel plots), and impedance spectrum (EIS) were collected using a
53 three-electrode system, with Si as the working electrode, standard
54 calomel electrode (SCE) as the reference, and platinum as the counter
55 electrode. Before each test, the Si electrode was sealed using insulating
56 tape, revealing a 1 cm² active area. Before each electrochemical
57 measurement, the samples were immersed in 5 wt% potassium hydroxide
58 solution for 5 min to remove the natural oxide layer. This was followed
59 by rinsing with deionized water and drying with compressed air. The
60 OCP was stabilized for around 600 seconds to ensure the acquisition of
61 trustworthy and consistent potential readings. Potentiodynamic
62 polarization was measured within a voltage range of OCP ± 0.3 V at a
63 scan rate of 5 mV/s. EIS was performed at the OCP, spanning a
64 frequency range of 0.01 Hz to 1000 kHz, with a 5 mV AC signal applied.
65 EIS data were analyzed and modeled using Zview software.

66 A uniform ink was prepared by dispersing 10 mg of the catalyst into
67 420 μL of a solution containing 200 μL of isopropanol, 200 μL of
68 deionized water, and 20 μL of Nafion solution, followed by 30 minutes of
69 ultrasonication. The resulting ink was evenly applied onto FTO glass,

70 with insulating tape to ensure exposure of a 1 cm² active area. Next, the
71 prepared electrode was annealed at 200°C for 2 hours.

72 The transient photocurrent response measurements were conducted
73 using a standard three-electrode system. Pt and Ag/AgCl electrodes
74 served as the counter and reference electrodes, respectively, and the
75 as-prepared electrode was used as the working electrode. Among these
76 measurements, the applied potential was 1 V vs. Ag/AgCl. The EIS
77 measurements were performed at the open circuit potential, ranging from
78 0.01 Hz to 1000 kHz. For these experiments, 0.5 mol/L KOH solution
79 was used as the electrolyte, and a 300 W Xe lamp served as the light
80 source.

81 The ORR measurements were conducted using a standard
82 three-electrode system, with Pt and Ag/AgCl electrodes serving as the
83 counter and reference electrodes, respectively, and the as-prepared
84 disk-ring electrode (5.61 mm in diameter, with an electrode surface area
85 of 0.2475 cm²) being used as the working electrode (Pine Research
86 Instrumentation). Linear sweep voltammetry (LSV) measurements were
87 recorded in an O₂-saturated 0.5 M KOH electrolyte solution with a scan
88 rate of 5 mV/s and a rotating speed of 1600 rpm. The potentials vs.
89 Ag/AgCl were converted to the RHE scale by the following equation:

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

90 In calculating the bandgap and DOS, the Geometry Optimization
91 adopt the DFT + U approach to improve the description of strong on-site
92 Coulomb repulsion of Ce 4 f electrons. U-J was set to 6.0 eV, which has
93 been widely reported to be effective. The cutoff energy of 400 eV was
94 used for all calculations, the k-point mesh of 4×4×4 was used, the energy
95 tolerance accuracy was set to 1×10⁻⁵ eV/atom, and the k-point was set to
96 8×8×8. The maximum force was selected at 0.03 eV/Å, and the

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

104

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

105 In calculating the electron density difference and surface O_v
106 formation energy, the Geometry Optimization of the experimental effect
107 of the GGA + U method with U = 5 eV for the reduction of CeO₂ under
108 UV irradiation for the Ce 4f state. A model of 16.199 Å was cleaved
109 using CeO₂ (111) as a section, from which supercells were built in the
110 range of $2 \times 2 \times 1$, and then a vacuum layer of 15 Å thickness was
111 created. The cutoff energy of 400 eV was used for all calculations, the
112 k-point mesh of $1 \times 1 \times 1$ was used, the energy tolerance accuracy was set
113 to 1×10^{-5} eV/atom, and the k-point was set to $2 \times 2 \times 2$. The maximum force
114 was selected at 0.02 eV/Å, and the maximum atom displacement was
115 placed at 1×10^{-3} Å. The calculations of electron density difference were
116 performed using a cutoff energy of 400 eV, for the Brillouin-zone
117 sampling, we used the $4 \times 4 \times 4$ Monkhorst-Pack mesh, and the
118 self-consistent convergence of the total energy is 2.0×10^{-6} eV/atom. The
119 O_v formation energy calculation is shown in the following equation:

120

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

121

122 **Characterization**

123 Raman spectra were recorded using the Horiba (Xplora Plus)
124 instrument. The contact angle is measured by a contact angle measuring
125 instrument (Powereach, JC2000D). The element concentration was
126 measured using a Thermo Scientific ICP-MS. Friction and temperature
127 were measured using GnP POLI-500 (G&P Technology, Inc). The
128 specific surface areas and pore size distribution were evaluated by the
129 BET method using N₂ adsorption (Micromeritics, ASAP 2460).
130 Hydrogen temperature-programmed reduction (H₂-TPR) and oxygen
131 temperature-programmed desorption (O₂-TPD) measurements were
132 performed using a BELCAT II system (MicrotracBEL, Japan). Thermal
133 imaging is captured by Infrared thermal imager (Fluke, Fluke-Ti300). For
134 H₂-TPR, the sample was pretreated under an argon flow (30 mL/min) by
135 heating from room temperature to 300 °C at a rate of 10 °C/min and
136 holding for 60 min. After cooling to 50 °C, a 10% H₂/Ar gas mixture
137 (50 mL/min) was introduced for 0.5 h. Once the baseline stabilized, the
138 temperature was increased to 800 °C at a rate of 10 °C/min under a 10%
139 H₂/Ar flow (30 mL/min). For O₂-TPD, the sample was similarly
140 pretreated in helium at 300 °C for 1 h. After cooling and stabilization, a
141 10% O₂/Ar gas mixture (50 mL/min) was introduced for 0.5 h. The
142 sample was then heated to 800 °C at 10 °C/min under a 10% O₂/Ar flow
143 (30 mL/min). Current–voltage (I–V) characteristics were measured using
144 an Agilent B1500A Semiconductor Device Analyzer in conjunction with
145 an EverBeing EB-8 probe station.

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

149 C1s peak (284.6 eV). The amount of Ce³⁺ 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:

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

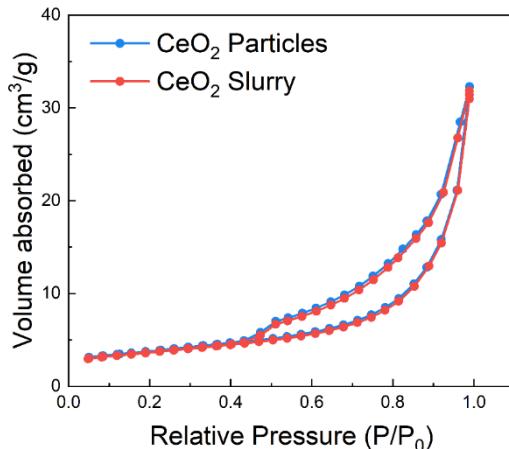
159 where A, h, ν, and α are constant values that refer to a constant, Plank
160 constant, light frequency and absorption coefficient, respectively.

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⁻¹ and energy changes
172 below 10⁻⁴ eV. Converged energy: E = -1.362572371481 × 10⁴ eV.

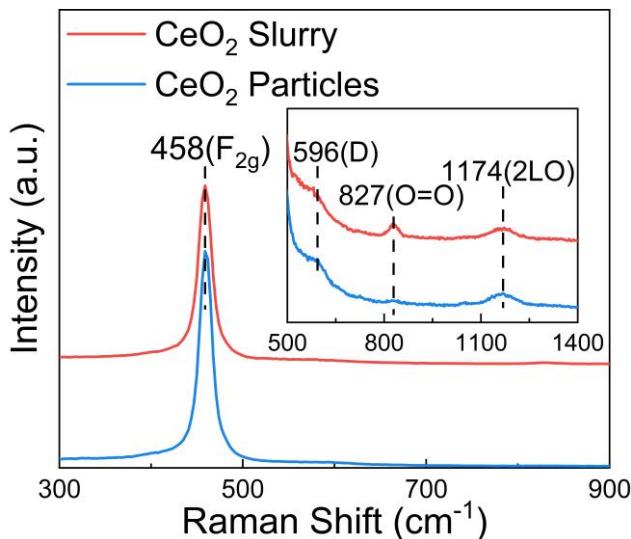
173 **Supplementary Figures**



174 **Supplementary Fig. S1.** BET surface area of CeO_2 particles and CeO_2 slurry.

175

176 **Discussion:** The specific surface areas of CeO_2 particles and CeO_2 slurry
177 were measured using the Brunauer–Emmett–Teller (BET). The specific
178 surface area of CeO_2 particles was $13.26 \text{ m}^2/\text{g}$, whereas that of the CeO_2
179 slurry was slightly lower, at $12.92 \text{ m}^2/\text{g}$. This minor reduction indicates
180 that the slurry preparation process has a negligible impact on the overall
181 surface area of the CeO_2 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.

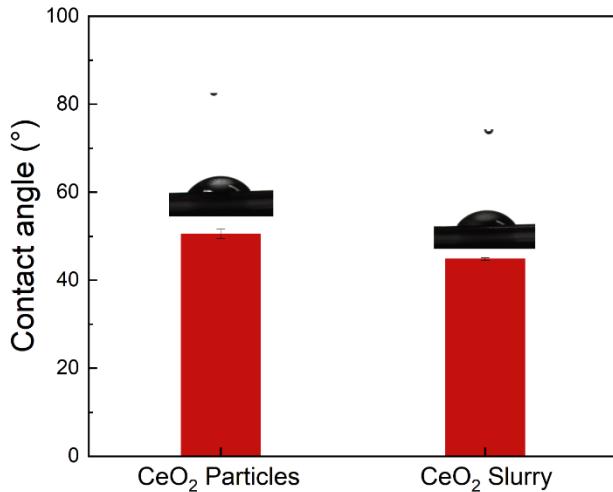


185 **Supplementary Fig. S2.** Raman spectrum, inset: the 500-1300 cm^{-1} shift region.

186

187 **Discussion:** The CeO_2 particles were primarily characterized by a peak at
 188 458 cm^{-1} , corresponding to the F_{2g} mode of the CeO_2 lattice¹. Additional
 189 peaks at 596 cm^{-1} , 827 cm^{-1} , and 1174 cm^{-1} were associated with
 190 defect-induced modes (D), adsorbed peroxy/superoxide species (O–O
 191 stretching), and longitudinal optical (2LO)². Adsorbed $\text{O}_2^{2-}/\text{O}_2^-$ species
 192 act as intermediates in the re-oxidation of reduced CeO_{2-x} to CeO_2 ³.
 193 Electrons donated by Ce^{3+} cations drive their conversion to Ce^{3+} .

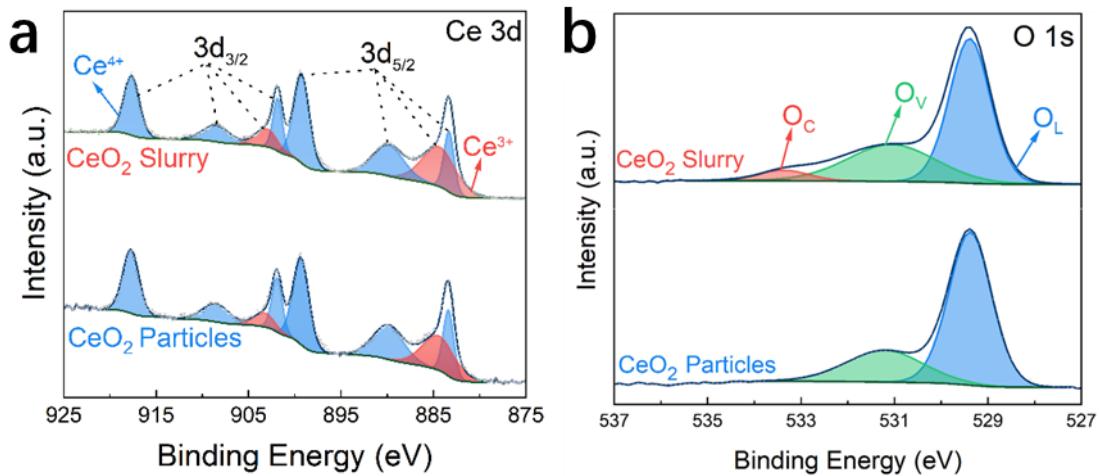
194 The ratio of the F_{2g} to defect peak intensities ($I_{\text{F}2g} / I_{\text{D}}$) serves as a
 195 sensitive measure of lattice order versus defectiveness. A higher ratio
 196 indicates better crystallinity and fewer defects, while a lower ratio reflects
 197 increased lattice disorder from oxygen vacancies, dopant effects, or other
 198 imperfections. In Fig. S2, these ratios are 44.16 for CeO_2 particles and
 199 38.64 for CeO_2 slurry, confirming a higher defect concentration in the
 200 slurry.



201 **Supplementary Fig. S3.** Contact angle of CeO₂ particles and CeO₂ slurry layer on Si
202 wafers surface with deionized water.

203

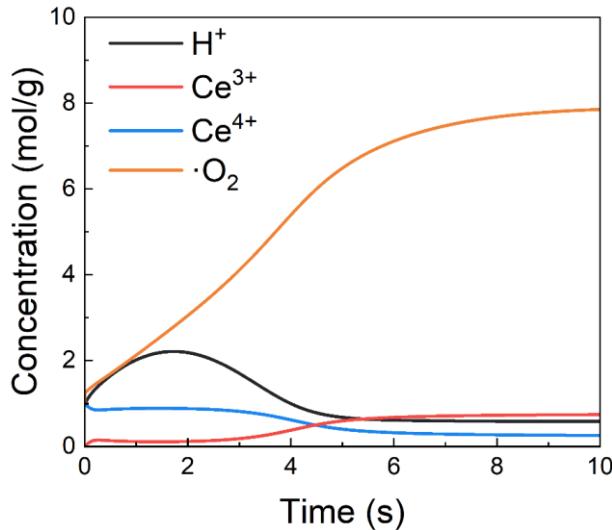
204 **Discussion:** Introducing oxygen vacancies leads to enhanced surface
205 charge accumulation, which reduces particle agglomeration and accounts
206 for the observed decrease in zeta potential. Moreover, oxygen vacancies
207 increase the surface free energy, promoting more effective interactions
208 between the CeO₂ abrasive and Si wafers. This effect is supported by
209 contact angle measurements, where the CeO₂ slurry demonstrated a
210 reduced contact angle of 44.56°, representing a decrease of 5.93°
211 compared to CeO₂ particles. This reduction is attributed to the increased
212 hydrophilicity resulting from water dissociative adsorption on the CeO₂
213 surface following the introduction of oxygen vacancies.



214 **Supplementary Fig. S4. a** Ce 3d, **b** O 1s XPS spectra of CeO₂ particles and CeO₂
 215 slurry.

216

217 **Discussion:** The Ce 3d spectra (Fig. S4a) comprise five spin-orbit split
 218 double peaks (3d_{3/2}, 3d_{5/2}), with 903.4 eV and 885.2 eV corresponding to
 219 Ce³⁺ signals⁴. Combined with the O 1s spectra (Fig. S4b), the results
 220 indicate that Janus CeO₂ slurry has a higher concentration of active sites.
 221 The relative contents of Ce³⁺ and oxygen vacancies are summarized in
 222 Table S1. The O 1s XPS spectrum shows peaks at 531.17 eV and 529.37
 223 eV, which are attributed to oxygen vacancies (O_V) and lattice oxygen
 224 (O_L), respectively, while the peak at 533.36 eV corresponds to
 225 chemisorbed oxygen species (O_C). Calculations indicate that the O_V
 226 density is 30.17% in CeO₂ particles and increases to 34.02% in the CeO₂
 227 slurry. The O_C originate from adsorbed peroxides (O₂²⁻/O₂⁻) on the CeO₂
 228 surface.

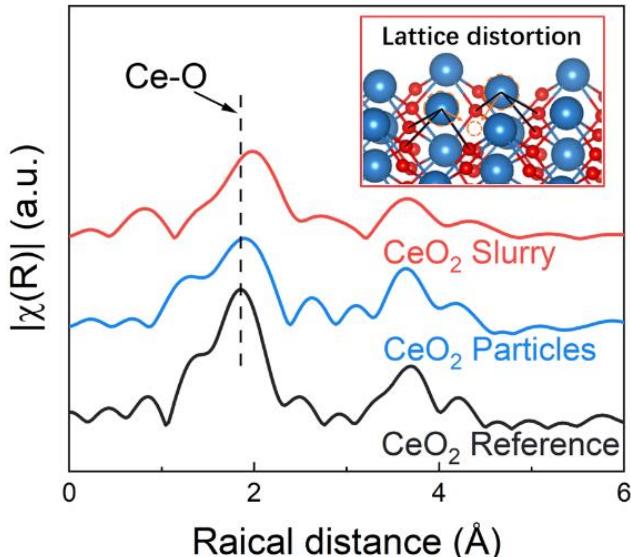


229 **Supplementary Fig. S5.** Disproportionation reaction of CeO_2 with H_2O_2 under
 230 alkaline conditions.

231

232 **Discussion:** During the photocatalytic CMP process, oxygen vacancy
 233 concentration of CeO_2 increases when CeO_2 is combined with H_2O_2 .
 234 Oxygen vacancies migrate to energetically favorable lattice positions, and
 235 Ce^{3+} relocates to the crystal surface. Due to the increased ionic radius,
 236 they are better accommodated spatially⁵. For this mechanism, a
 237 reasonable model for the superoxide disproportionation reaction
 238 mechanism at the cerium dioxide nanoparticle surface⁶.

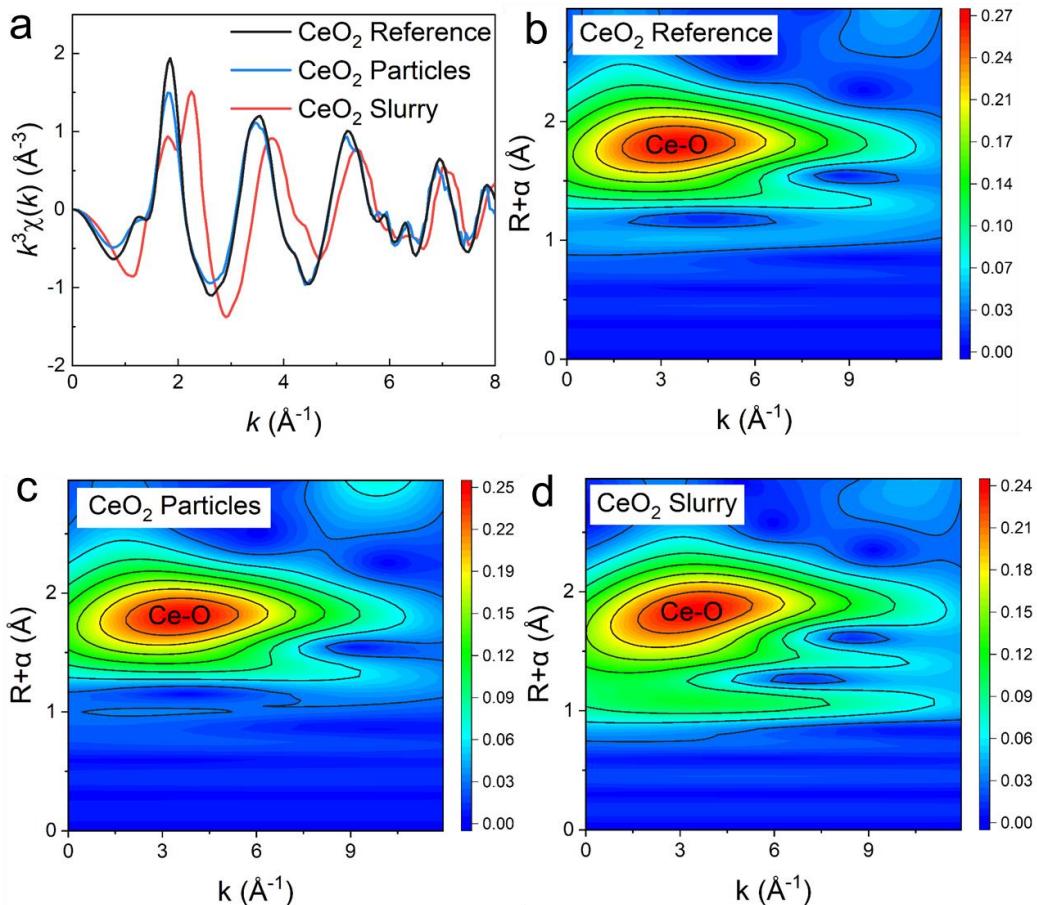
239 The CeO_2 slurry shows alkalinity. Therefore, the Runge-Kutta
 240 method was used for modeling. Under alkaline conditions, the conversion
 241 between Ce^{3+} and Ce^{4+} occurs and eventually reaches equilibrium. When
 242 hydrogen peroxide is added, Ce^{4+} actively controls the superoxide,
 243 resulting in a minor transformation of Ce^{3+} ions, leading to oxygen
 244 vacancies.



245 **Supplementary Fig. S6.** Radial distribution function from EXAFS, inset: lattice
 246 distortion and coordination environment change in CeO_2 slurry.

247

248 **Discussion:** The R-space spectra and fitting data (Table S2) further reveal
 249 variations in the Ce-O bond length within the CeO_2 slurry. Introducing
 250 oxygen vacancies results in the elongation of first-shell Ce-O bonds,
 251 leading to surface lattice distortion. These findings are further supported
 252 by the K-space EXAFS fitting spectra and wavelet transform analysis.



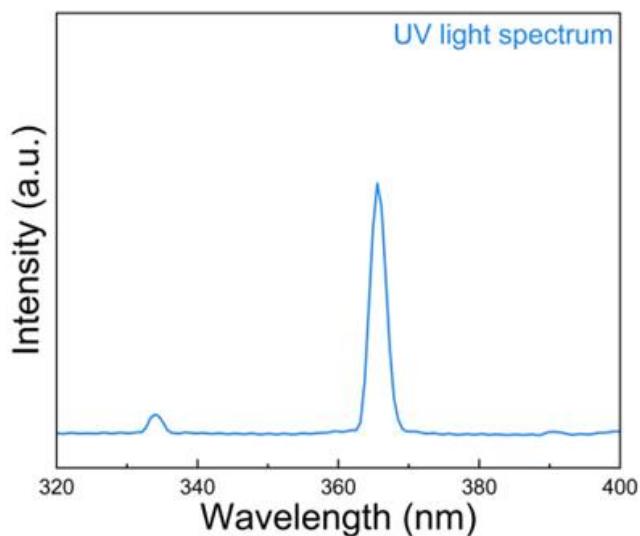
253 **Supplementary Fig. S7.** **a** The corresponding K space EXAFS fitting spectra of
 254 CeO₂ reference, CeO₂ particles, and CeO₂ slurry. Wavelet transform of Ce K-edge
 255 EXAFS spectra for **b** CeO₂ reference, **c** CeO₂ particles, and **d** CeO₂ slurry.

256

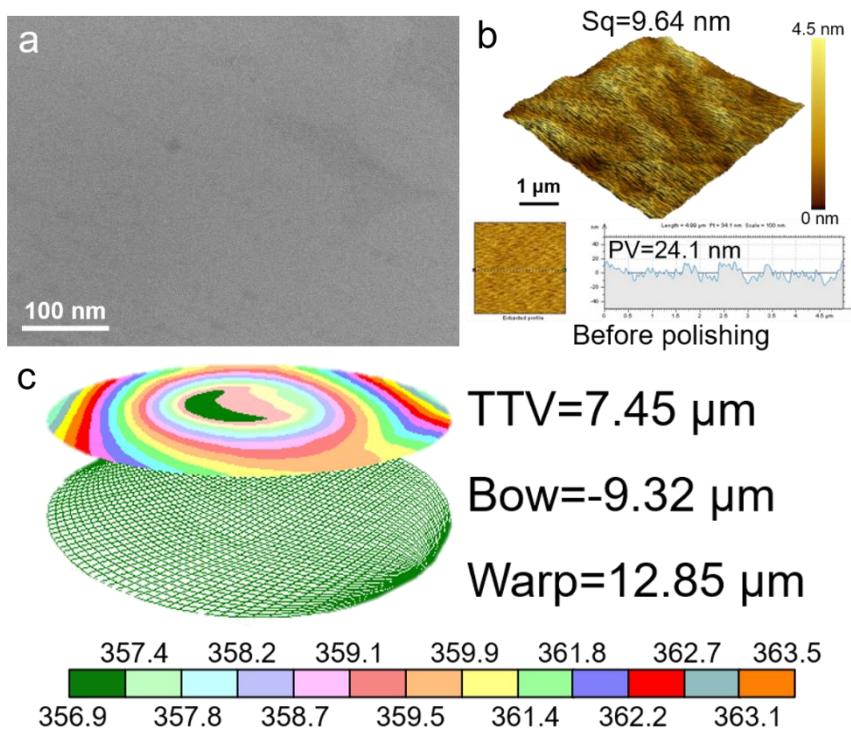
257 **Discussion:** Combining the results of the EXAFS fits (Table S2) reveals
 258 a lower coordination number and a higher Debye-Waller factor for the
 259 CeO₂ slurry, suggesting a more disordered surface structure. Additionally,
 260 oxygen vacancies partially contribute to the Ce-O-Ce shell, thereby
 261 promoting greater electron delocalization.

262 The local chemical environment and physical structure around the
 263 Ce atoms were investigated using Ce K-edge EXAFS spectra. From the
 264 wavelet transform contour plots (Fig. S7a), each sample exhibited distinct
 265 maxima attributed to the scattering effect of neighboring O atoms (Ce-O
 266 bonding, first shell, $k \sim 3.6 \text{ \AA}^{-1}$). Compared to the CeO₂ reference and

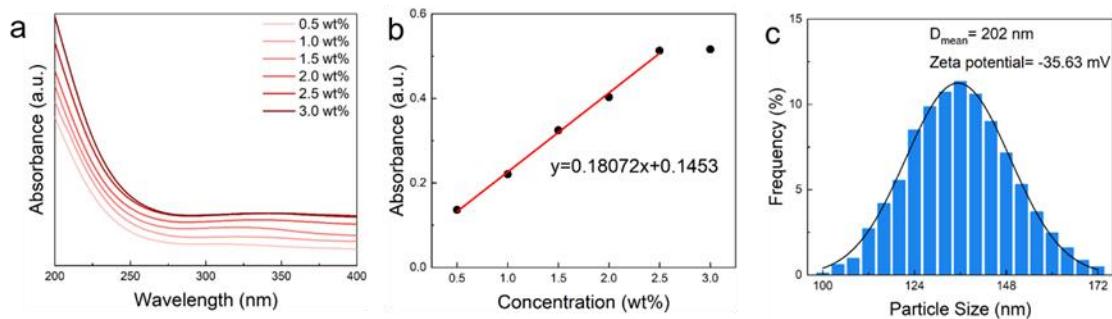
267 CeO₂ particles, the intensity of Ce-O bonding was significantly reduced
268 for the CeO₂ slurry, indicating a higher concentration of oxygen
269 vacancies. This reduction is attributed to homogeneous interfaces that
270 disrupt the Ce atom arrays, inducing lattice distortions and suppressing
271 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.



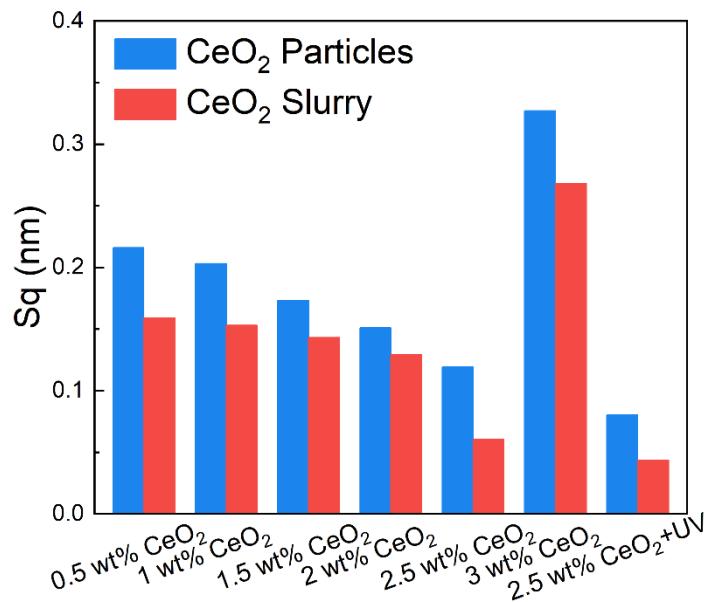
275 **Supplementary Fig. S9.** **a** 3D AFM image and height profiles. **b** SEM image. **c**
 276 Geometric parameter image of Si CMP using CeO_2 particles.



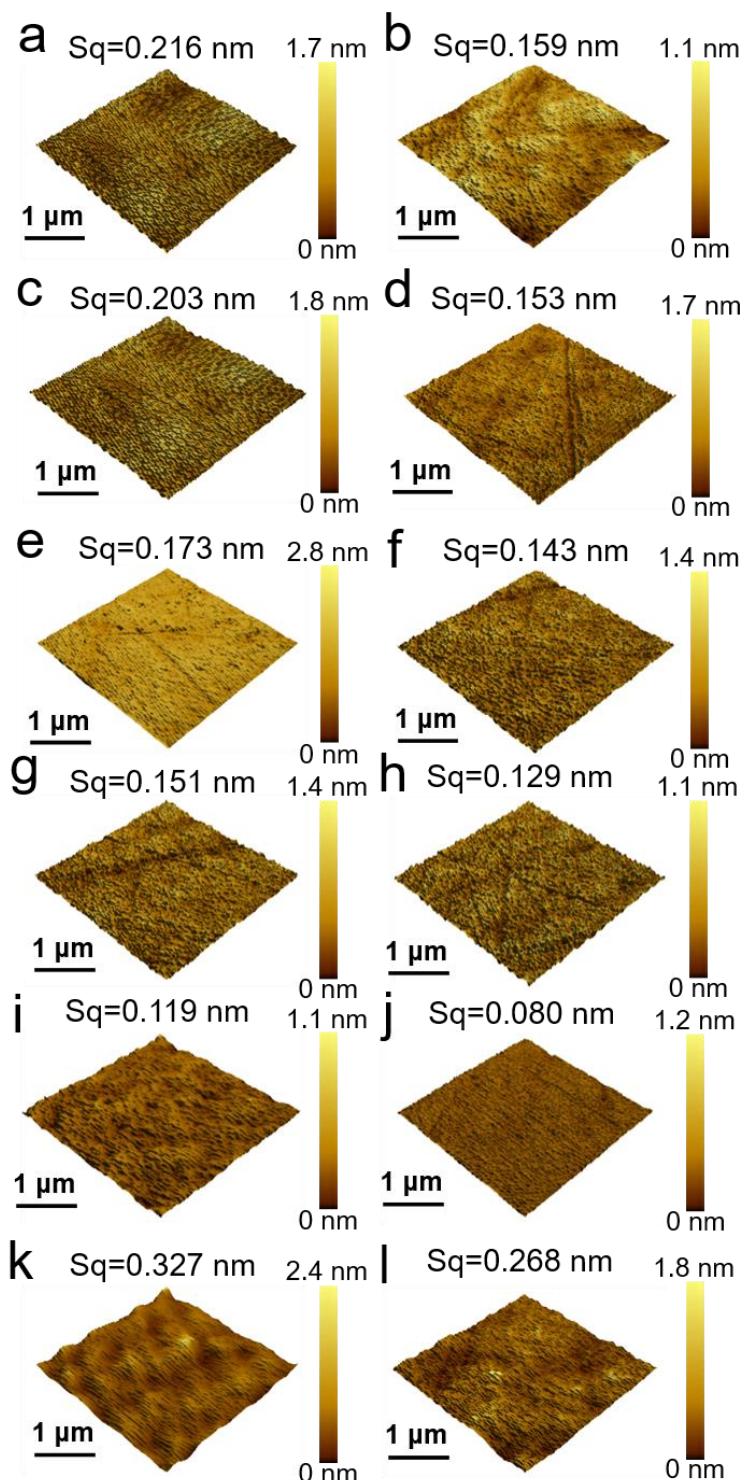
277 **Supplementary Fig. S10.** (a) UV-Vis absorption spectra of CeO₂ slurry of different
 278 concentrations. (b) This fit shows a linear trend with a slope of 0.18072 called the
 279 exponent coefficient. (c) Particle size distribution histogram and zeta potential of 3
 280 wt.% CeO₂, which exhibits an average CeO₂ particle size of 136.5 nm and zeta
 281 potential of -35.63 mV.

282

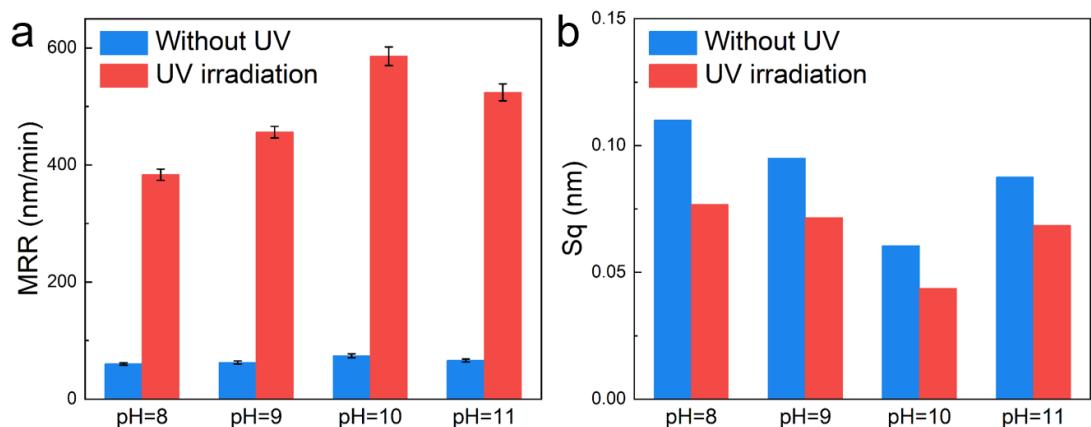
283 **Discussion:** The intensity of the characteristic absorption peak gradually
 284 increased, with the CeO₂ slurry reaching maximum absorbance at a
 285 concentration of 2.5 wt.%, indicating that the CeO₂ abrasives were
 286 approaching saturation in the dispersion. However, upon further
 287 increasing the CeO₂ concentration, the abrasives exceeded saturation,
 288 resulting in particle aggregation and sedimentation. This supersaturation
 289 led to a decrease in both the dispersion quality and the stability of the
 290 polishing solution.



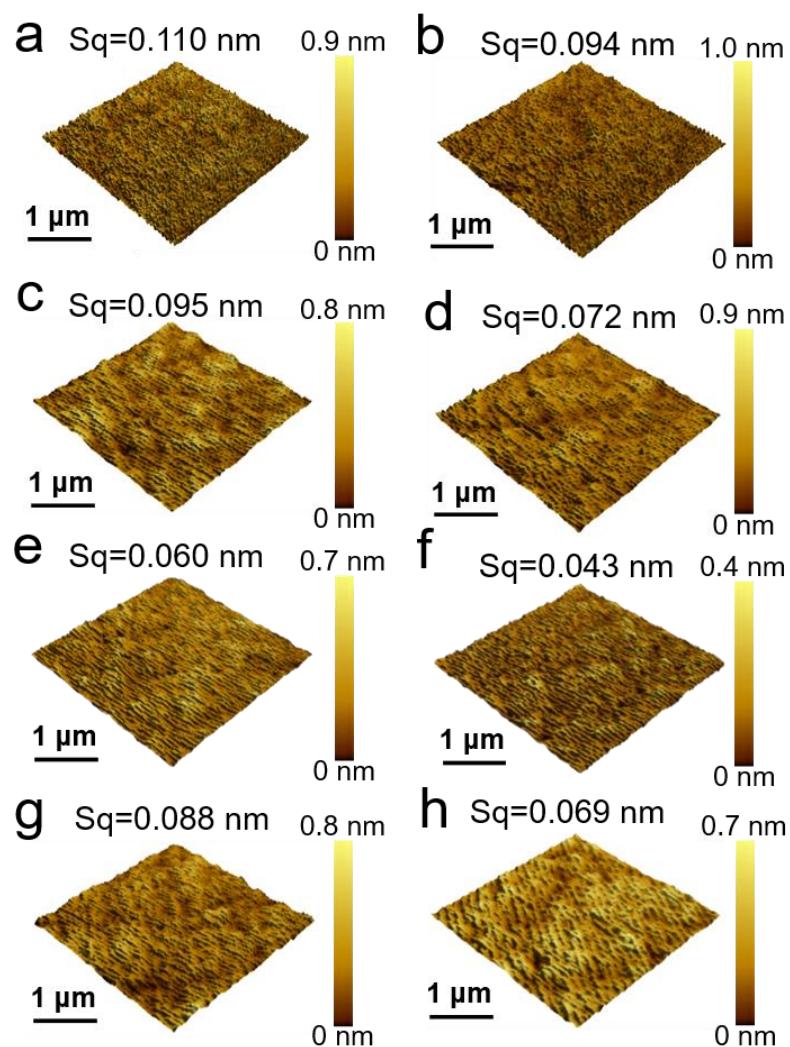
291 **Supplementary Fig. S11.** S_q comparison for CeO_2 particles and Janus CeO_2 slurry at
292 various weight concentrations and UV illumination.



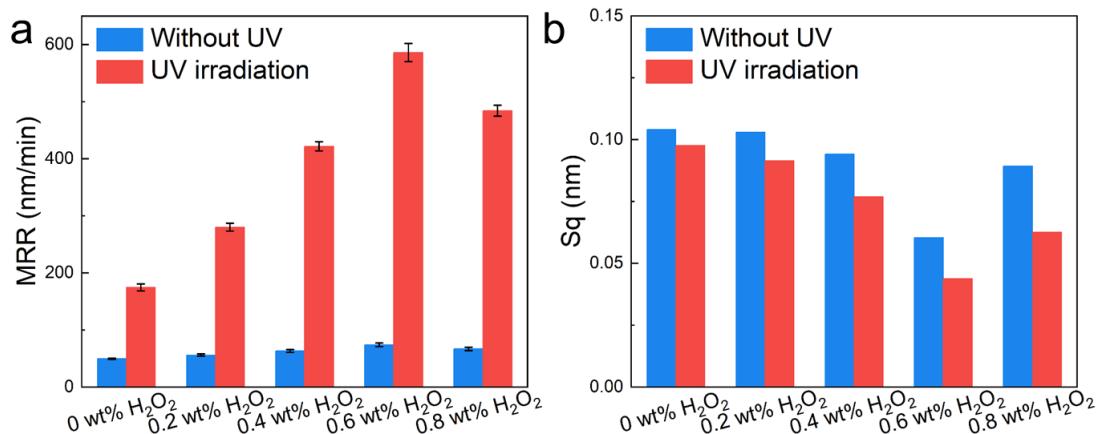
293 **Supplementary Fig. S12.** 3D AFM images of Si wafer surfaces polished under
 294 various weight concentrations CeO₂ particles and Janus CeO₂ slurry: **a** 0.5 wt%
 295 (particles); **b** 0.5 wt% (slurry); **c** 1 wt% (particles); **d** 1 wt% (slurry); **e** 1.5 wt%
 296 (particles); **f** 1.5 wt% (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).



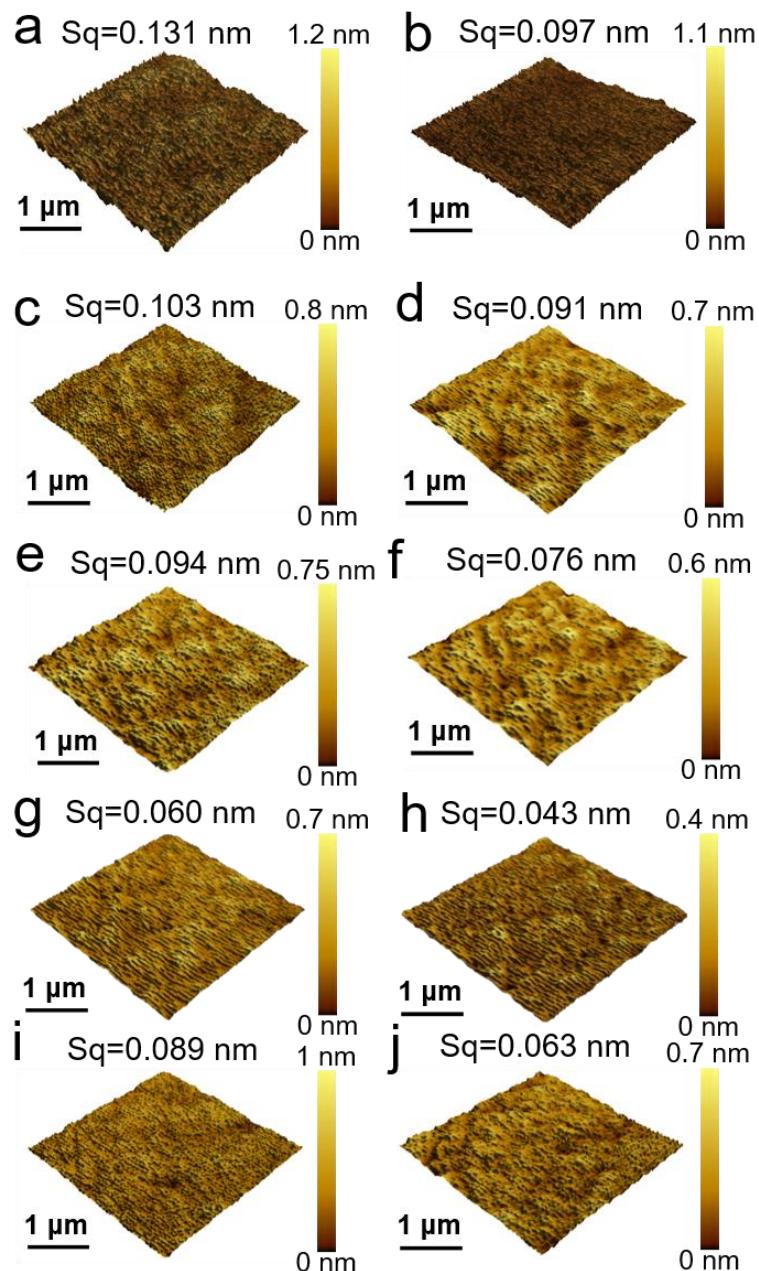
298 **Supplementary Fig. S13.** Comparison of **a** MRR, **b** Sq for Si wafers polished at
299 different pH levels.



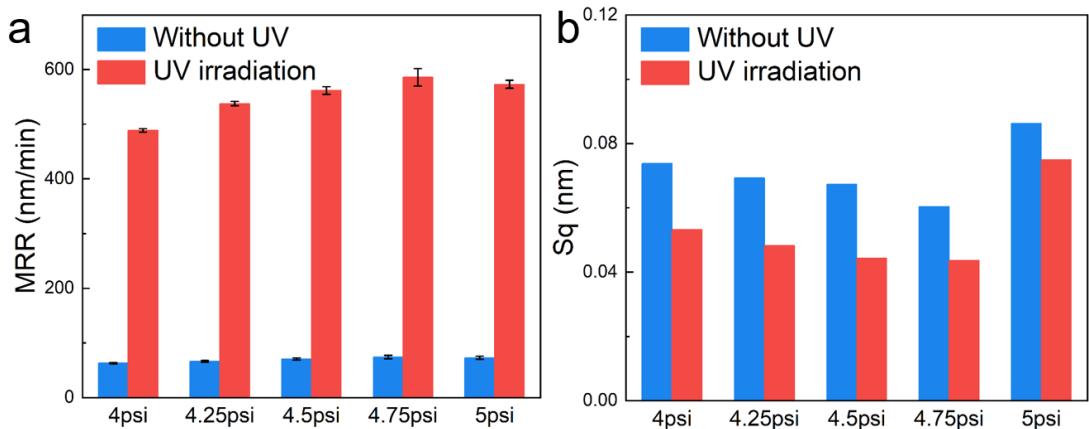
300 **Supplementary Fig. S14.** 3D AFM images of Si wafer surfaces polished under
 301 different pH: **a** pH=8; **b** pH=8 (UV); **c** pH=9; **d** pH=9 (UV); **e** pH=10; **f** pH=10 (UV);
 302 **g** pH=11; **h** pH=11 (UV).



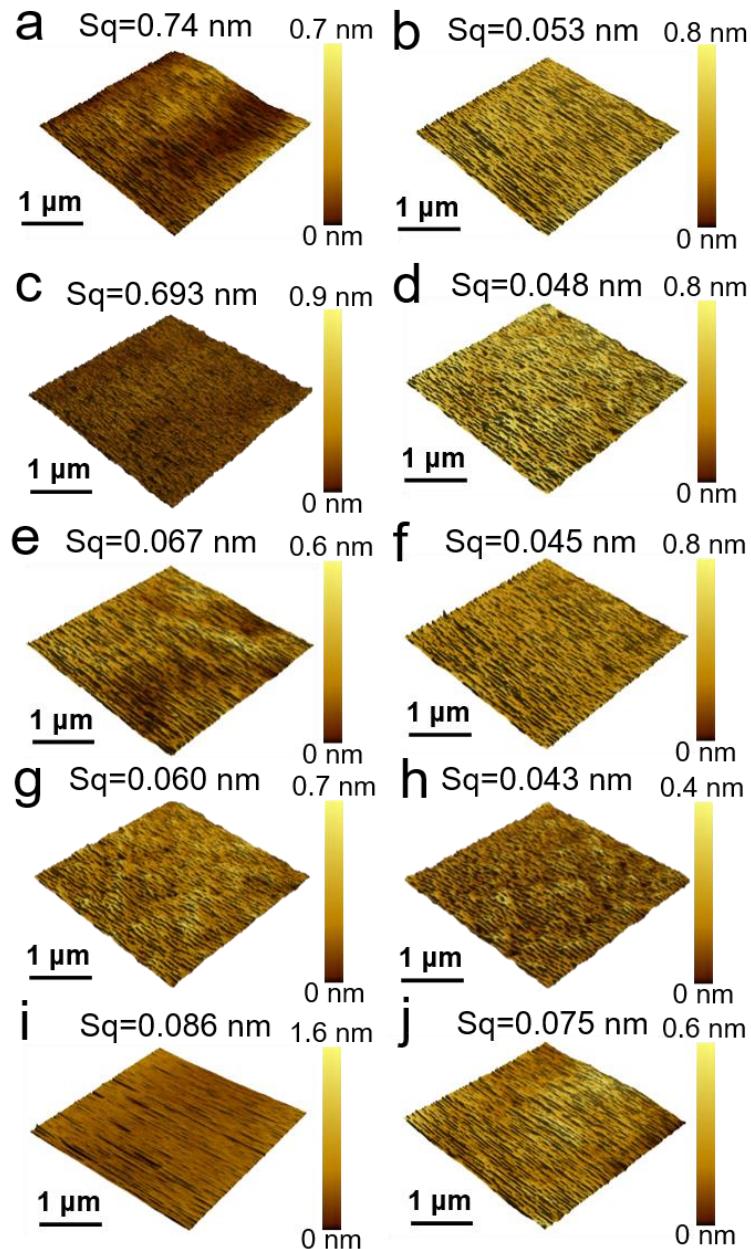
303 **Supplementary Fig. S15.** Comparison of **a** MRR, **b** S_q for the Si wafers polished
 304 under different weight concentrations of H_2O_2 .



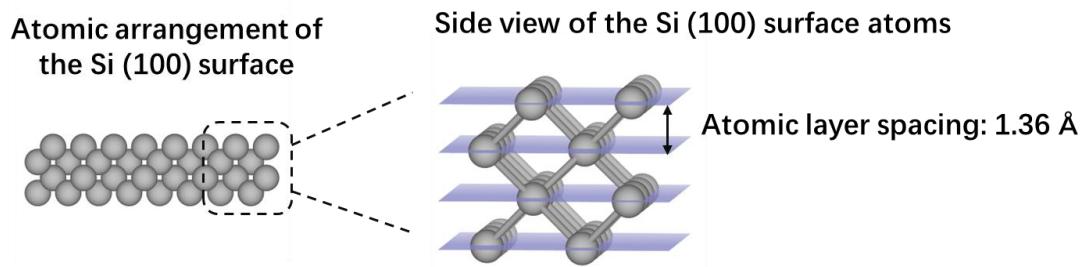
305 **Supplementary Fig. S16.** 3D AFM images of Si wafer surfaces polished under
 306 different weight concentrations of H_2O_2 : **a** 0 wt%; **b** 0 wt% (UV); **c** 0.2 wt%; **d** 0.2
 307 wt% (UV); **e** 0.4 wt%; **f** 0.4wt% (UV); **g** 0.6 wt%; **h** 0.6 wt% (UV); **i** 0.8 wt%; **j** 0.8
 308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



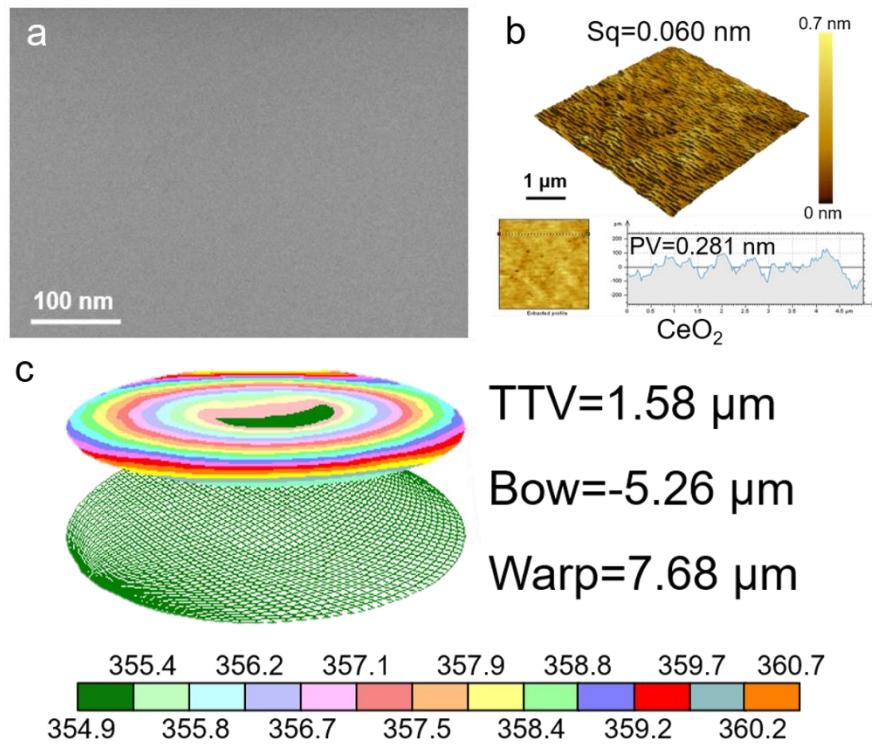
309 **Supplementary Fig. S17.** Comparison of **a** MRR, **b** Sq on the Si wafers polishing
310 under different polishing pressure.



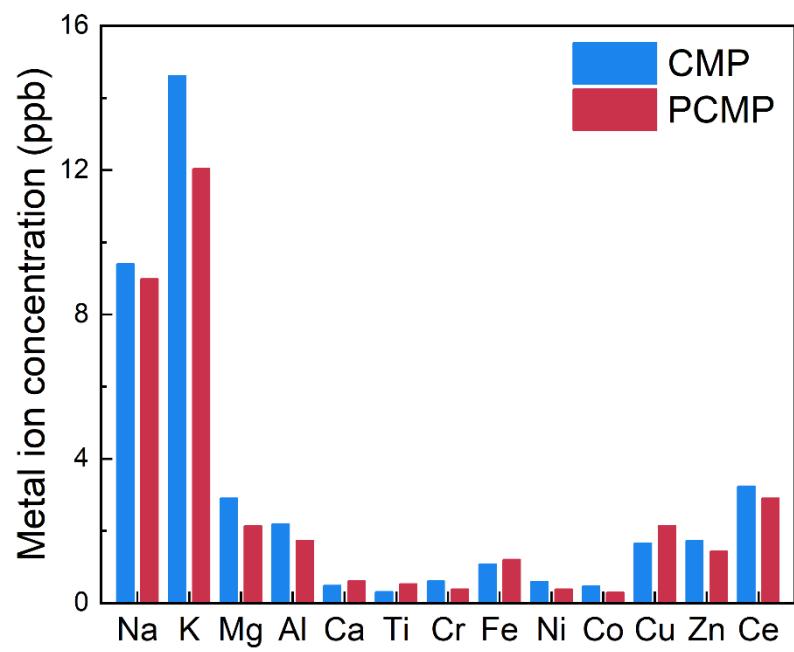
311 **Supplementary Fig. S18.** 3D AFM images of Si wafer surfaces polished under
 312 different polishing pressure: **a** 4 psi; **b** 4 psi (UV); **c** 4.25 psi; **d** 4.25 psi (UV); **e** 4.5
 313 psi; **f** 4.5 psi (UV); **g** 4.75 psi; **h** 4.75 psi (UV); **i** 5 psi; **j** 5 psi (UV).



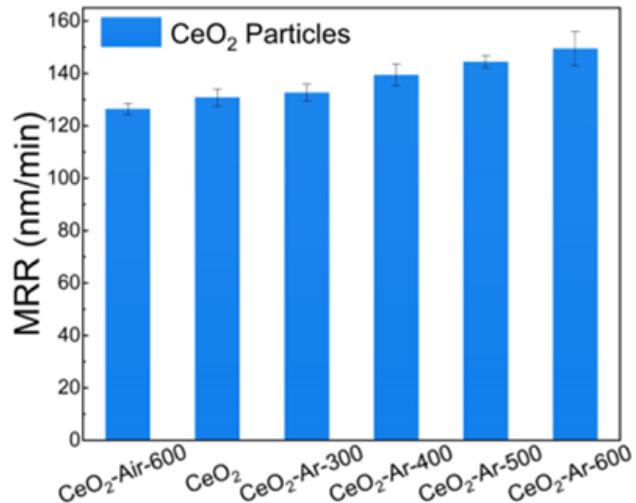
314 **Supplementary Fig. S19.** Atomic arrangement of the Si (100) surface.



315 **Supplementary Fig. S20.** **a** 3D AFM image and height profiles. **b** SEM image. **c** Si
 316 Geometric parameter image using Janus CeO_2 slurry (2.5 wt%).



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

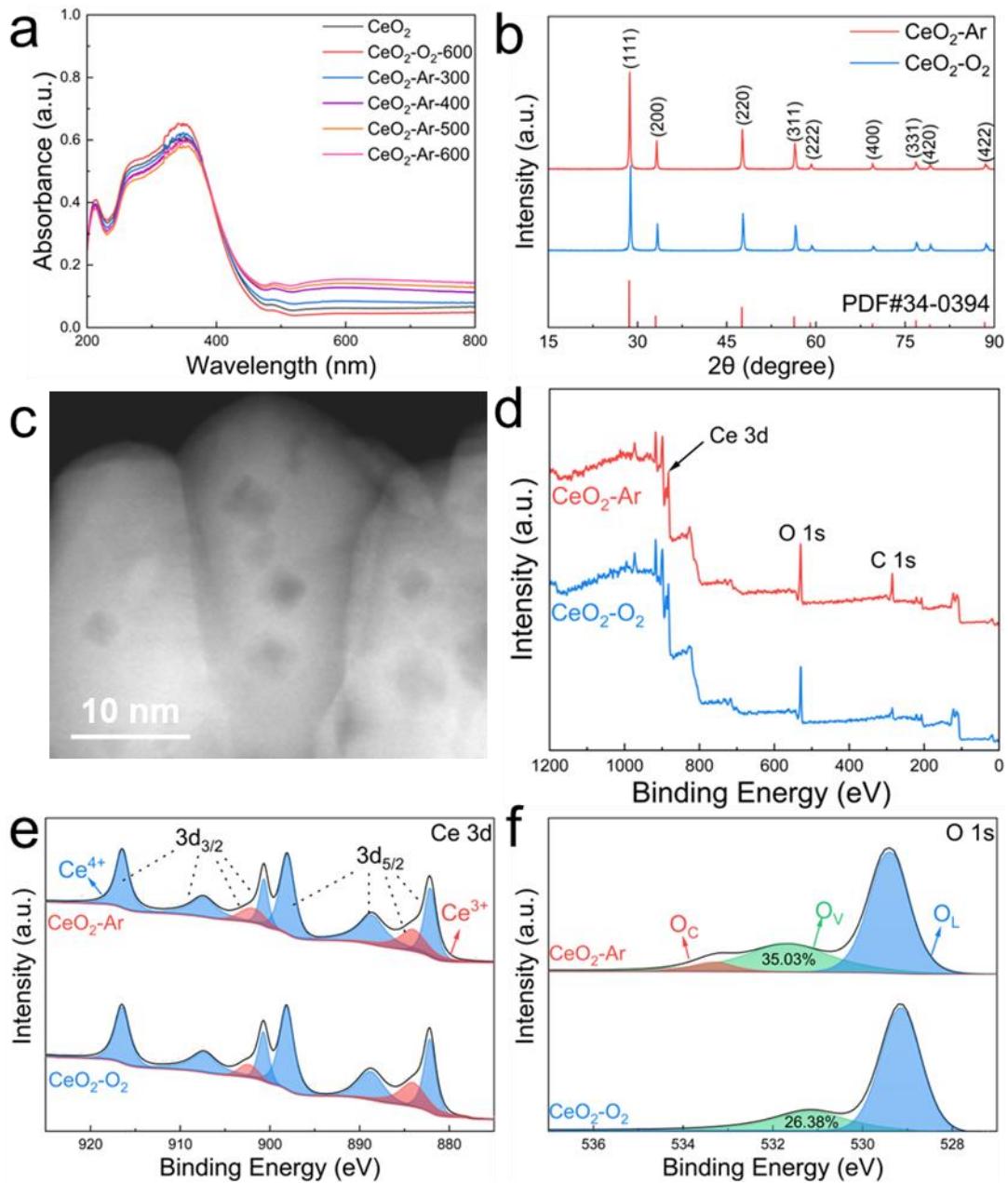


318 **Supplementary Fig. S22. a** Comparison of MRR on SiO₂ wafers with CeO₂ under
 319 different conditions.

320

321 **Discussion:** The MRR of SiO₂ during CMP using CeO₂ particles
 322 subjected to different calcination conditions was investigated. Samples
 323 calcined in air (CeO₂-Air-600) exhibited the lowest MRR, attributed to
 324 the reduced concentration of oxygen vacancies, as air calcination tends to
 325 replenish lattice oxygen. In contrast, CeO₂ samples calcined in an Ar
 326 atmosphere showed progressively higher MRRs with increasing
 327 calcination temperatures (300–600 °C), due to the enhanced formation of
 328 oxygen vacancies. These results indicate that oxygen vacancies in CeO₂
 329 can improve its chemical reactivity, thereby promoting more efficient
 330 SiO₂ removal during CMP.

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

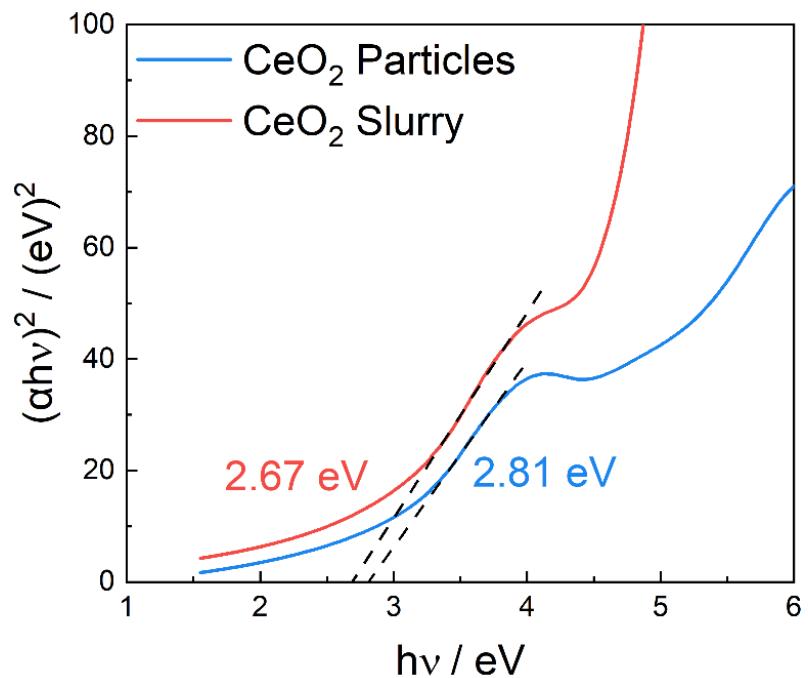


337 **Supplementary Fig. S23.** **a** UV-Vis spectra. **b** XRD pattern. **c** HAADF-STEM of
 338 CeO₂-Ar-600. **d** XPS spectra, **e** Ce 3d, **d** O 1s XPS spectra of CeO₂-Ar and CeO₂-O₂.
 339
 340 **Discussion:** Oxygen treatment tends to fill oxygen vacancies, while
 341 annealing in Ar atmosphere promotes oxygen diffusion out of the lattice,
 342 thereby generating oxygen vacancies. As shown in Fig. S23a, visible light
 343 absorption in the 500–800 nm range gradually increases with the

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

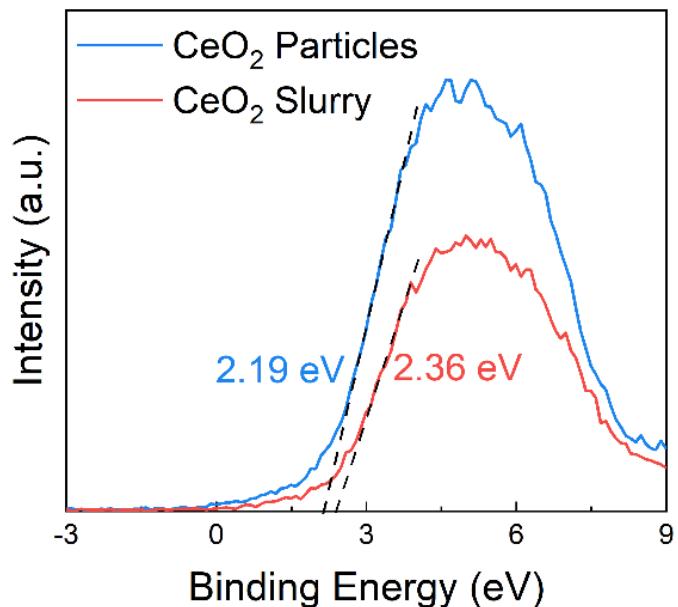
346 XRD analysis (Fig. S23b) reveals a leftward shift of diffraction
347 peaks in CeO_2 samples annealed under Ar ($\text{CeO}_2\text{--Ar}$) compared to those
348 treated in O_2 ($\text{CeO}_2\text{--O}_2$), consistent with lattice expansion. This shift is
349 attributed to two coupled effects: (1) the formation of oxygen vacancies,
350 which locally disrupt the fluorite structure, and (2) the partial reduction of
351 Ce^{4+} to Ce^{3+} , whose larger ionic radius (1.14 Å for Ce^{3+} vs. 0.97 Å for
352 Ce^{4+} in 8-fold coordination) leads to elongation of Ce–O bonds to
353 preserve charge neutrality⁷.

354 These structural modifications are further corroborated by
355 HAADF-STEM and XPS (Fig. S23c–f). The HAADF-STEM image
356 clearly displays abundant surface defects on $\text{CeO}_2\text{--Ar}$ particles. XPS
357 analysis of the O 1s spectra shows that the oxygen vacancy contribution
358 increases from 26.38% in $\text{CeO}_2\text{--O}_2$ to 35.03% in $\text{CeO}_2\text{--Ar}$ (Fig. S23f),
359 validating the effectiveness of Ar annealing in enhancing defect
360 concentration. Together, these results demonstrate a strong correlation
361 between reduction state, lattice parameter, and oxygen vacancy
362 concentration, highlighting the critical role of defect thermodynamics in
363 tailoring the structural and electronic properties of ceria-based materials.



364

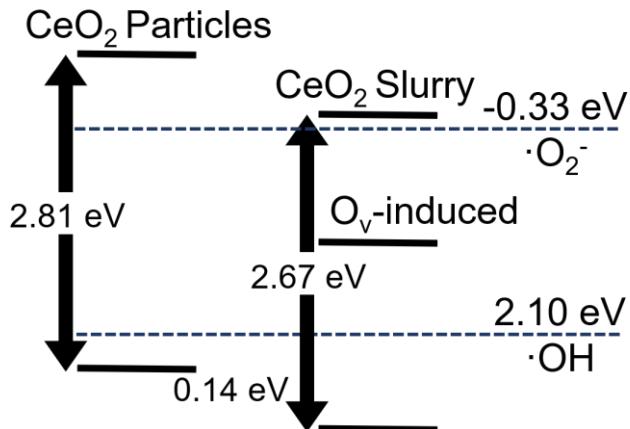
Supplementary Fig. S24. Tauc plot of CeO_2 particles and CeO_2 slurry.



365 **Supplementary Fig. S25.** UPS valence band spectra of CeO₂ particals and slurry.

366

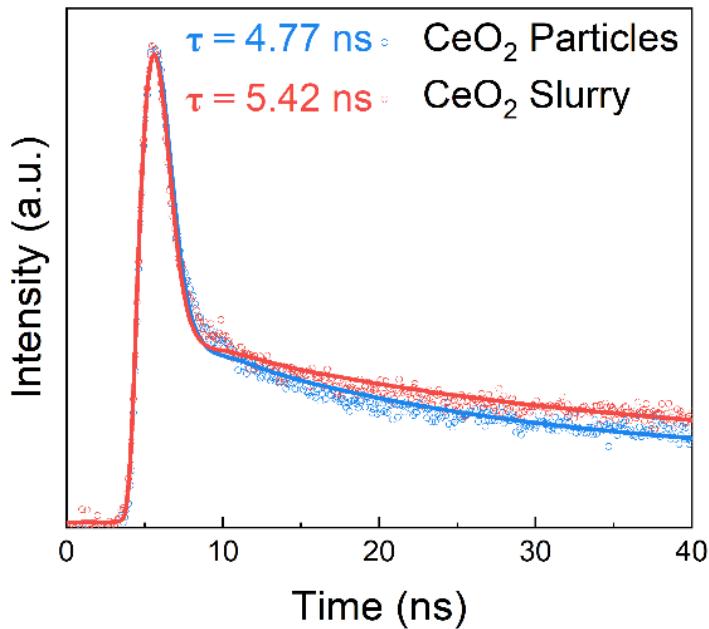
367 **Discussion:** The presence of oxygen vacancies causes electrons at the top
368 of the valence band to shift to lower energy levels⁸, resulting in valence
369 band potentials of 2.36 eV for CeO₂ particles and 2.19 eV for Janus CeO₂
370 slurry.



371 **Supplementary Fig. S26.** The band structure of CeO_2 particles and CeO_2 slurry.

372

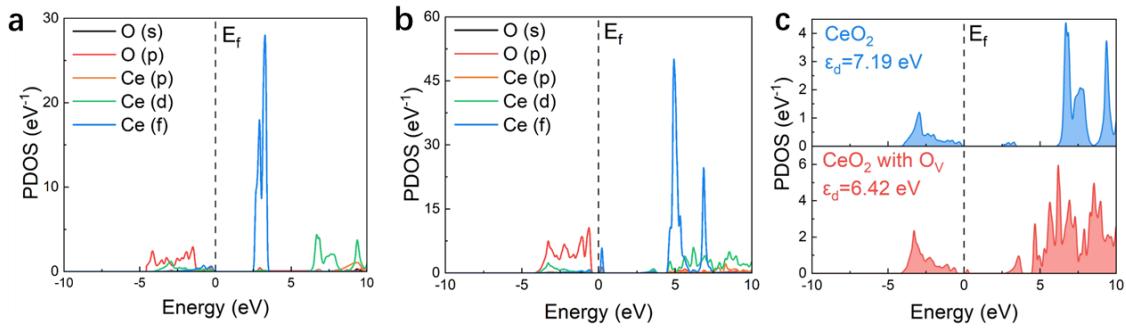
373 **Discussion:** The constructed band structure reveals that the Janus CeO_2
 374 slurry possesses a band gap of 2.67 eV, corresponding to an absorption
 375 edge of approximately 465 nm, which enables efficient excitation under
 376 365 nm UV irradiation. Notably, the conduction band minimum (CBM)
 377 lies above the redox potential of $\text{O}_2/\cdot\text{O}_2^-$ (-0.33 V vs. NHE), while the
 378 valence band maximum (VBM) is below the redox potential of $\text{H}_2\text{O}/\cdot\text{OH}$
 379 (+2.38 V vs. NHE). This favorable band alignment facilitates the
 380 generation of both superoxide ($\cdot\text{O}_2^-$) and hydroxyl ($\cdot\text{OH}$) radicals upon
 381 photoexcitation. These reactive oxygen species contribute synergistically
 382 to the oxidative enhancement observed during the PCMP process.



383 **Supplementary Fig. S27.** Time-resolved PL decay curves of CeO_2 particles and
384 CeO_2 slurry.

385

386 **Discussion:** The transient fluorescence lifetimes of CeO_2 particles and
387 Janus CeO_2 slurry are 4.77 ns and 5.42 ns, respectively. It indicates
388 oxygen vacancies act as an electronic trap to induce local band structure
389 changes in CeO_2 , particularly forming mid-gap states in regions with
390 oxygen deficiency^{9, 10}. These mid-gap states act as intermediate trapping
391 sites for electrons transitioning from the excited to the ground state,
392 slowing the electron recombination pathway and thereby extending the
393 lifetime of photo-generated charge carriers.

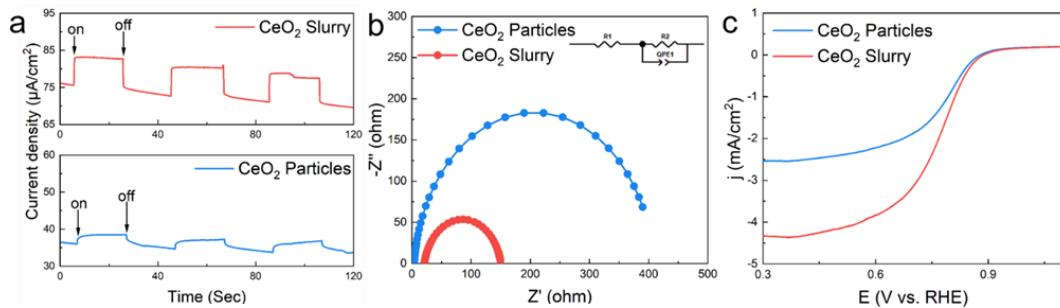


394 **Supplementary Fig. S28.** **a** Projected density of states of CeO_2 . **b** CeO_2 with O_v . **c**
 395 Projected density of states of Ce 3d orbitals. The Fermi level is set to zero.

396

397 **Discussion:** The electronic structures of CeO_2 with and without oxygen
 398 vacancies were analyzed by partial density of states (PDOS) calculations.

399 It reveals that oxygen vacancies lead to excess electrons occupying the
 400 Ce 4f states, creating new electronic states, and the excess electron gain
 401 by the 4f would give rise to the charge carrier in the band. Compared to
 402 CeO_2 , the d-band center of CeO_2 with oxygen vacancy is closer to the
 403 0.77 eV. Therefore, defect states cause band tailing and act as mid-states,
 404 enabling electrons to be excited from the valence band to the conduction
 405 band through multi-step excitation processes. This enhances the light
 406 utilization efficiency of CeO_2 and boosts the generation rate and lifetimes
 407 of photo-generated carriers.

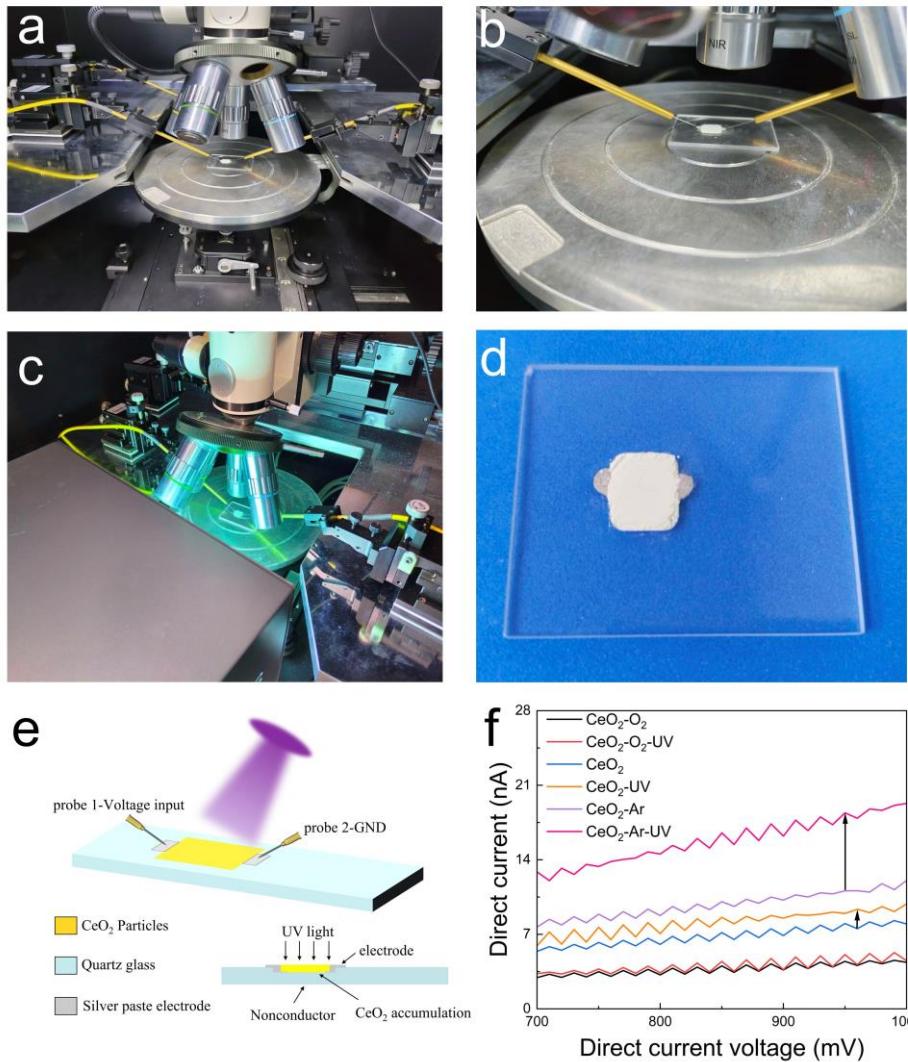


408 **Supplementary Fig. S29.** **a** Transient photocurrent response. **b** Polarization curves. **c**
 409 Nyquist plot of CeO₂ particles and CeO₂ slurry.

410

411 **Discussion:** The photocurrent densities over time under three cycles of
 412 intermittent illumination (Fig. S29a) demonstrated that CeO₂ particles
 413 exhibited weak current densities when illuminated, whereas the CeO₂
 414 slurry produced a distinct photocurrent response. Upon switching off the
 415 Xe lamp, the photocurrent values of both samples rapidly declined.
 416 Notably, the photocurrent density of the CeO₂ slurry reached 7.55
 417 μA/cm², which was 3.07 times higher than that of CeO₂ particles.

418 In the Nyquist plot (Fig. S29b), the semicircular radius for the CeO₂
 419 slurry was smaller than that of CeO₂ particles, indicating lower charge
 420 transfer resistance. Additionally, the CeO₂ slurry displayed superior
 421 oxygen reduction reaction (ORR) performance in linear sweep
 422 voltammetry (LSV) measurements (Fig. S29c), with an onset potential of
 423 0.87 V and a half-wave potential (E_{1/2}) of 0.81 V, outperforming the CeO₂
 424 particles (0.77 V and 0.74 V, respectively). These results highlight the
 425 highly efficient photogenerated charge transport capabilities of the CeO₂
 426 slurry compared to CeO₂ particles.

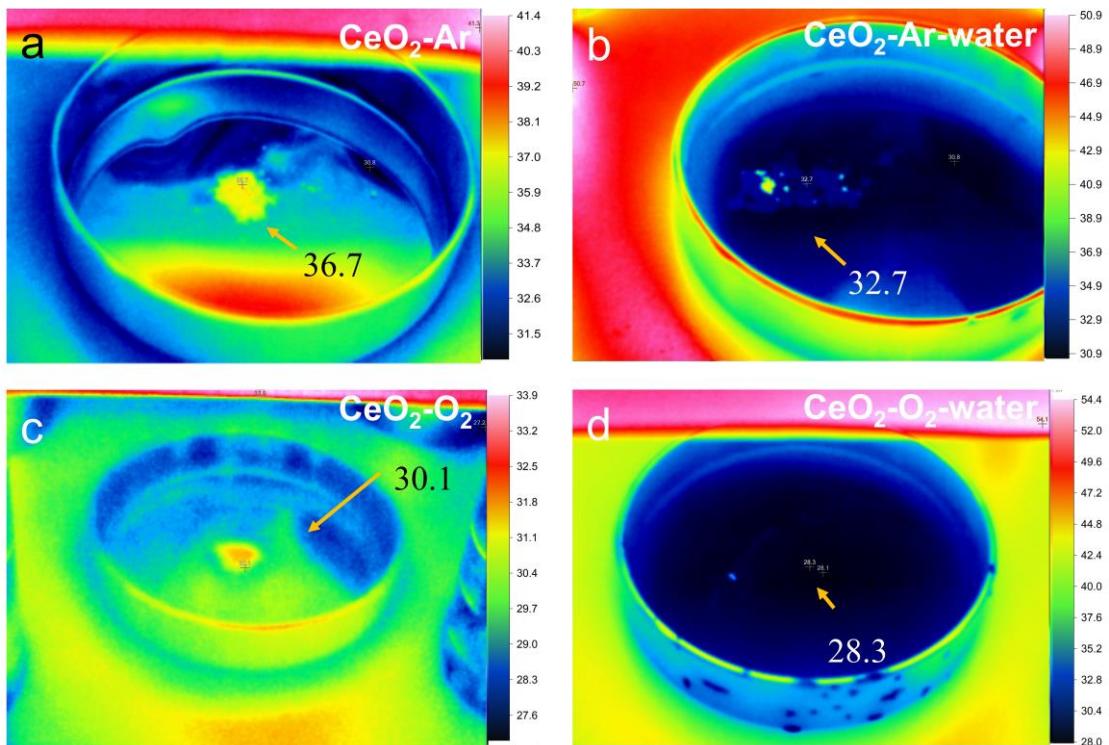


427 **Supplementary Fig. S30.** **a** Schematic illustration of the I-V sweep of
 428 electrochemical electrode. **b** Probe contacting. **c** Electrochemical measurements under
 429 UV light. **d** Electrode containing cerium dioxide particles. **e** Schematic illustration of
 430 the electrochemical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

432

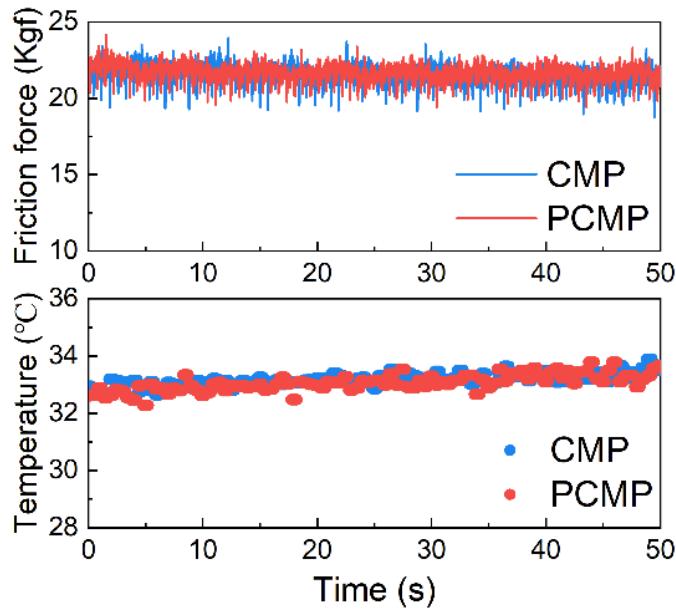
433 **Discussion:** The photoresponse behavior of various CeO₂ were evaluated
 434 under UV irradiation using a two-probe I-V measurement setup (Fig.
 435 S30a-e). The samples were deposited on quartz glass substrates with
 436 silver paste electrodes, and UV light was applied vertically to excite
 437 photo-generated carriers. As shown in Fig. S30f, the CeO₂ sample

438 calcined under an Ar atmosphere and subjected to UV irradiation
439 (CeO_2 –Ar–UV) exhibited the highest photocurrent among all samples.
440 This enhanced photocurrent is attributed to the high concentration of
441 oxygen vacancies, which introduce shallow defect levels within the band
442 gap. These defect levels act as trapping centers for photo-generated
443 electrons, reducing carrier recombination and facilitating charge
444 separation and transport. Additionally, the localized states enable
445 sub-bandgap excitation and improved conductivity under UV light. In
446 contrast, CeO_2 calcined in an O_2 atmosphere exhibited much lower
447 photocurrent, due to the fill of oxygen vacancies, which hinders the
448 excitation and transport of photo-generated carriers.



449 **Supplementary Fig. S31.** Infrared thermal images of CeO_2 samples under UV
 450 illumination. **a** $\text{CeO}_2\text{-Ar}$, **b** $\text{CeO}_2\text{-Ar-water}$, **c** $\text{CeO}_2\text{-O}_2$, and **d** $\text{CeO}_2\text{-O}_2\text{-water}$.

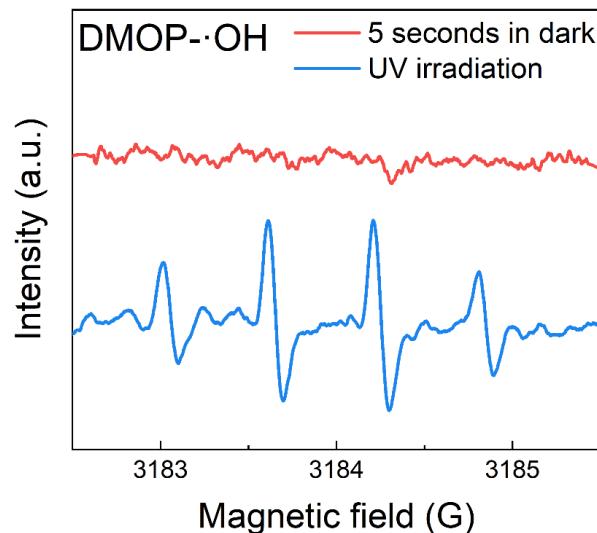
451
 452 **Discussion:** The $\text{CeO}_2\text{-Ar}$ sample exhibits the highest surface
 453 temperature ($36.7\text{ }^\circ\text{C}$), attributed to the presence of abundant oxygen
 454 vacancies that promote non-radiative recombination and local heating. In
 455 contrast, $\text{CeO}_2\text{-O}_2$, with fewer oxygen vacancies, shows a significantly
 456 lower temperature ($30.1\text{ }^\circ\text{C}$). The addition of water reduces the surface
 457 temperature due to its cooling and shielding effects, with $\text{CeO}_2\text{-O}_2\text{-water}$
 458 reaching the lowest temperature ($28.3\text{ }^\circ\text{C}$). These results highlight the role
 459 of oxygen vacancies and medium composition in regulating photothermal
 460 behavior.



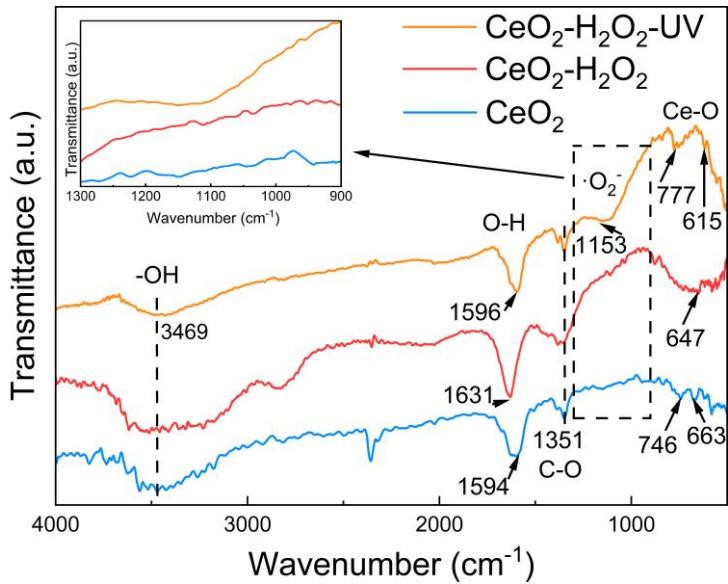
461 **Supplementary Fig. S32.** Friction force and slurry temperature during conventional
 462 CMP and PCMP.

463

464 **Discussion:** As shown in Fig. S32, the friction force and temperature
 465 remain relatively stable and comparable between CMP and PCMP,
 466 indicating similar polishing conditions. This is because the slurry is in
 467 dynamic flow during polishing, preventing heat accumulation in the bulk
 468 and limiting temperature rise to the CeO_2 surface. The localized heating
 469 of CeO_2 surface originates from non-radiative recombination of
 470 photo-generated carriers on the CeO_2 surface, which releases energy as
 471 phonons and elevates the temperature at the abrasive–wafer interface.



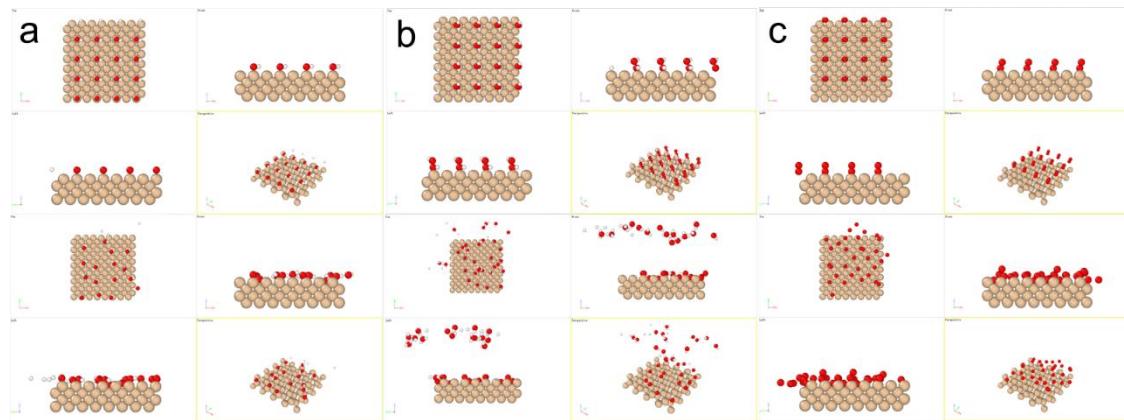
472 **Supplementary Fig. S33.** ESR spectra of DMPO-·OH for CeO₂ slurry under
473 different conditions.



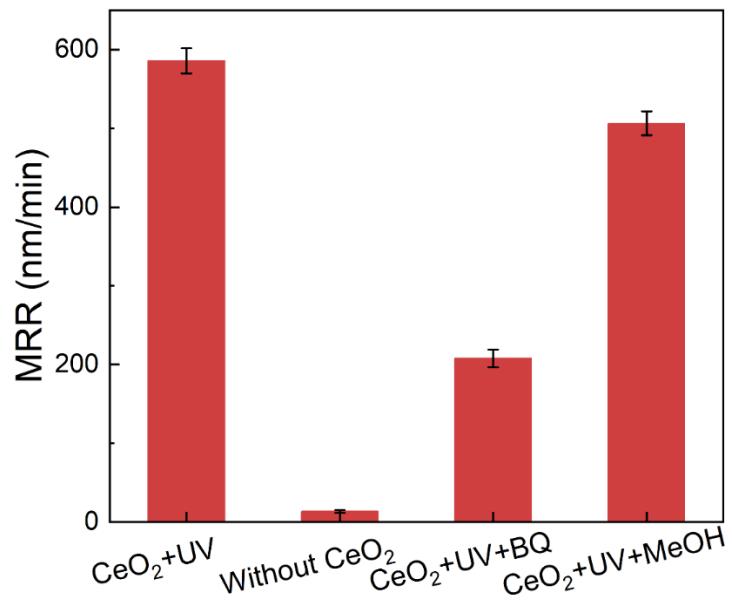
474 **Supplementary Fig. S34.** FTIR of CeO_2 under different conditions.

475

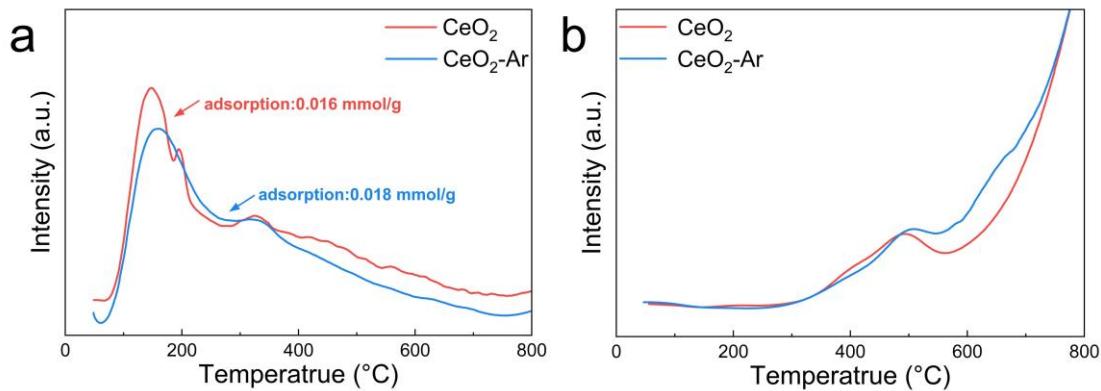
476 **Discussion:** A drop of H_2O_2 was added onto the pressed CeO_2 film,
477 followed by UV irradiation after a 2-minute. Compared to pristine CeO_2 ,
478 the $\text{CeO}_2\text{--H}_2\text{O}_2$ and $\text{CeO}_2\text{--H}_2\text{O}_2\text{--UV}$ samples exhibit several new or
479 enhanced peaks, indicating chemical interactions between CeO_2 and H_2O_2 .
480 The broad absorption band around 3469 cm^{-1} is assigned to $-\text{OH}$
481 stretching vibrations, while peaks near 1631 cm^{-1} and 1596 cm^{-1}
482 correspond to O-H bending modes. A prominent peak at 1351 cm^{-1} is
483 attributed to C-O stretching. After UV irradiation, a new band appears at
484 1153 cm^{-1} , which is assigned to the formation of superoxide species on
485 the CeO_2 surface. The Ce-O lattice vibrations are observed in the lower
486 wavenumber region ($900\text{--}500\text{ cm}^{-1}$), with shifts from 663 cm^{-1} and
487 746 cm^{-1} to 647 cm^{-1} and 777 cm^{-1} in the UV-treated sample, suggesting
488 lattice distortion and defect formation. The inset highlights the
489 enhancement in the $900\text{--}1300\text{ cm}^{-1}$ region, further confirming the
490 generation of reactive oxygen species¹¹.



491 **Supplementary Fig. S35.** Molecular dynamic simulation of silicon oxidation with
492 (a) $\cdot\text{OH}$, (b) H_2O_2 , and (c) $\cdot\text{O}_2^-$, with a molecular number of 16.



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

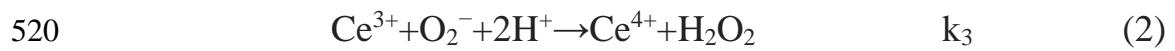
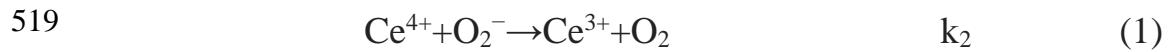


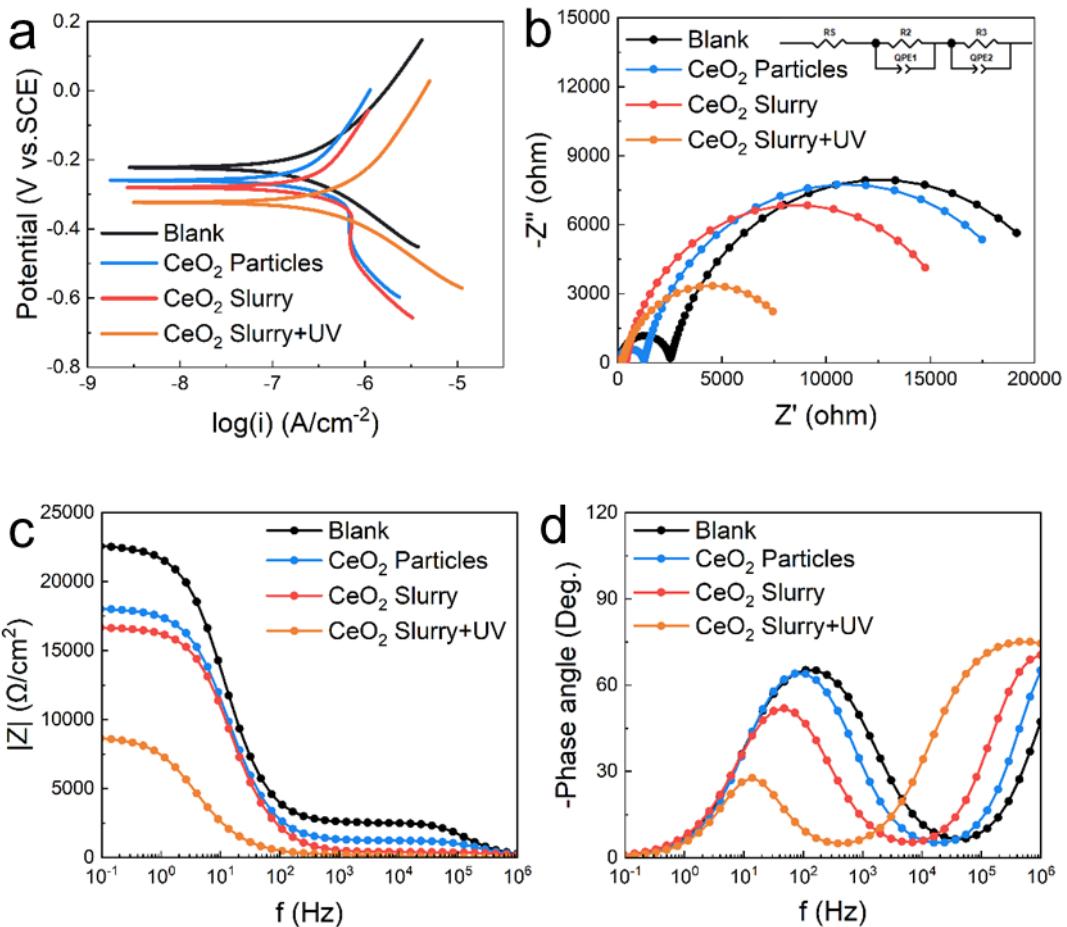
495 **Supplementary Fig. S37.** (a) O₂-TPD profiles of CeO₂, CeO₂-Ar-600 showing the
 496 adsorption amounts and strength of O₂. (b) H₂-TPR profiles of CeO₂ materials
 497 showing the reducibility of reactive SOS.

498

499 **Discussion:** Since TPD and TPR measurements require pretreatment to
 500 remove surface contaminants, and the Janus CeO₂ slurry contains organic
 501 components such as surfactants, which can interfere with the evaluation
 502 of oxygen vacancy adsorption and desorption behavior, Ar-calcined CeO₂
 503 was used for characterization. Surface oxygen species on oxide
 504 nanomaterials include surface lattice oxygen atoms as well as adsorbed
 505 reactive oxygen species such as *O₂, *O₂⁻, and *O₂²⁻. The influence of
 506 enhanced defect states on O₂ adsorption and activation was investigated
 507 using O₂-temperature programmed desorption (O₂-TPD), which identifies
 508 the types and strengths of oxygen adsorption. For CeO₂ particles,
 509 desorption of O₂ between 50–200 °C is attributed to weakly adsorbed
 510 *O₂⁻, while *O₂ desorbs between 150–300 °C¹². As shown in Fig. S37a,
 511 compared to CeO₂, oxygen-deficient CeO₂ exhibits weaker physical
 512 adsorption but stronger chemical adsorption between 50–250 °C, based
 513 on acidity comparisons¹³. Fig. S37b further demonstrates that CeO₂ with
 514 oxygen vacancies possesses a higher O₂ storage capacity, consistent with
 515 the higher values of kinetic constants k₂ and k₃, facilitating the
 516 subsequent formation of superoxide radicals. Additionally, the desorption

517 curve at elevated reduction temperatures indicates that CeO₂-Ar-600
518 retains the oxidation capability associated with surface oxygen species.





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

525

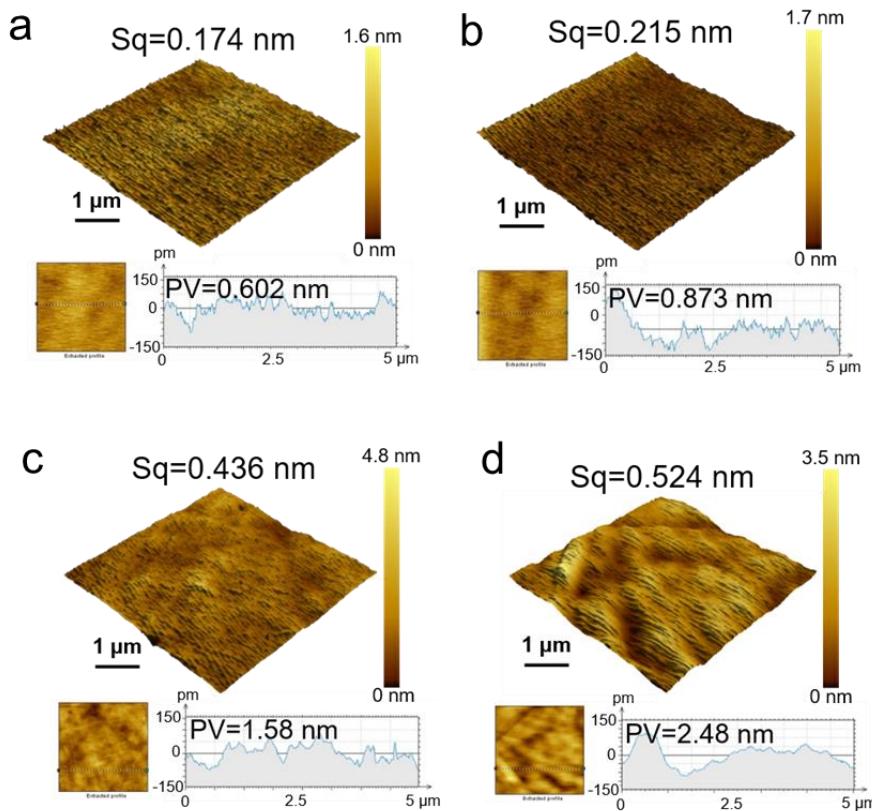
526 **Discussion:** To further explore the contributions of various slurry
 527 components and the influence of UV light, the oxidation processes of the
 528 Si surface were systematically analyzed using potential dynamic
 529 polarization curves (Fig. S38a). The corrosion potential (E_{corr}) is the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

534 current density (I_{corr}) quantifies the rate at which corrosion proceeds. It is
535 determined from the Tafel extrapolation of the polarization curve and is
536 directly proportional to the corrosion rate. A higher I_{corr} means a faster
537 corrosion rate, indicating that the material is undergoing more rapid
538 electrochemical degradation¹⁴.

539 The E_{corr} and I_{corr} , derived from Tafel plots, are summarized in Table
540 S3. In the dispersed system of CeO₂ particles, the self-catalytic reaction
541 requires significant interfacial pressure for initiation, resulting in an E_{corr}
542 of -0.222 V and I_{corr} of 6.561×10^{-7} A/cm². With the successive addition
543 of slurry components and the introduction of UV light, E_{corr} and I_{corr}
544 progressively increased, eventually reaching -0.323 V and 2.067×10^{-6}
545 A/cm². This progressive increase reflects the intensification of
546 UV-induced photocatalytic reactions, which significantly accelerate the
547 oxidation of the Si wafer.

548 Electrochemical Impedance Spectroscopy (EIS) can probe the
549 interfacial properties of materials by applying a small-amplitude AC
550 voltage over a wide range of frequencies and measuring the resulting
551 current. To further understand the interfacial dynamics, EIS parameters
552 were analyzed using a fitted equivalent circuit model (Fig. S38b), with
553 impedance values compiled in Table S4. The interfacial charge transfer
554 resistance decreased through the interaction between UV light and CeO₂,
555 reaching $R1 = 215 \Omega \cdot \text{cm}^2$. This reduction could be attributed to
556 generating electron-hole pairs that altered the ionic distribution at the
557 interface between CeO₂ slurry and Si wafer under UV illumination,
558 thereby enhancing the double-layer capacitance (QPE1) to 2.923×10^{-8}
559 nF/cm². The increased capacitance decreased the low-frequency
560 impedance, highlighting the improved interfacial charge transfer between
561 CeO₂ and Si.

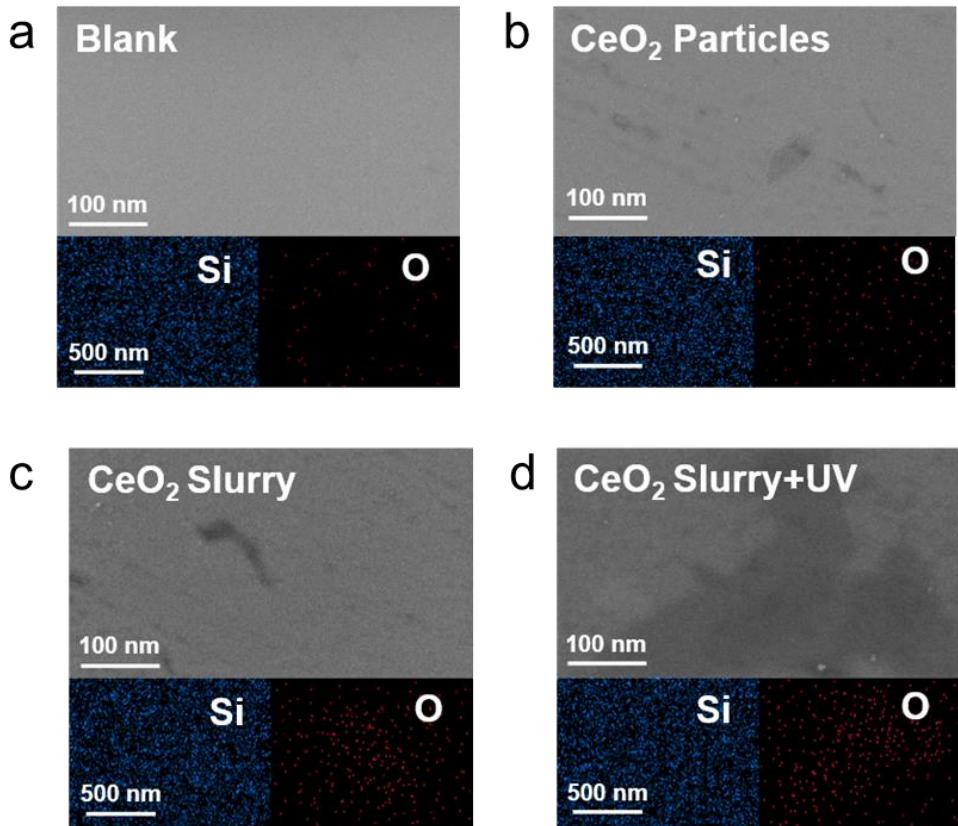
562 The amplitude and phase Bode plots further corroborate this trend.
563 Upon introducing UV light, the amplitude Bode plot (Fig. S38c)
564 demonstrated a marked decrease in impedance modulus ($|Z|$) within the
565 low-frequency region, suggesting a reduction in charge transfer resistance
566 (R_{ct}). 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.



571 **Supplementary Fig. S39.** The 3D AFM images with corresponding height profiles
 572 for **a** Si wafer before immersion, **b** CeO₂ particles, **c** CeO₂ slurry, and **d** CeO₂ slurry
 573 with UV illumination.

574

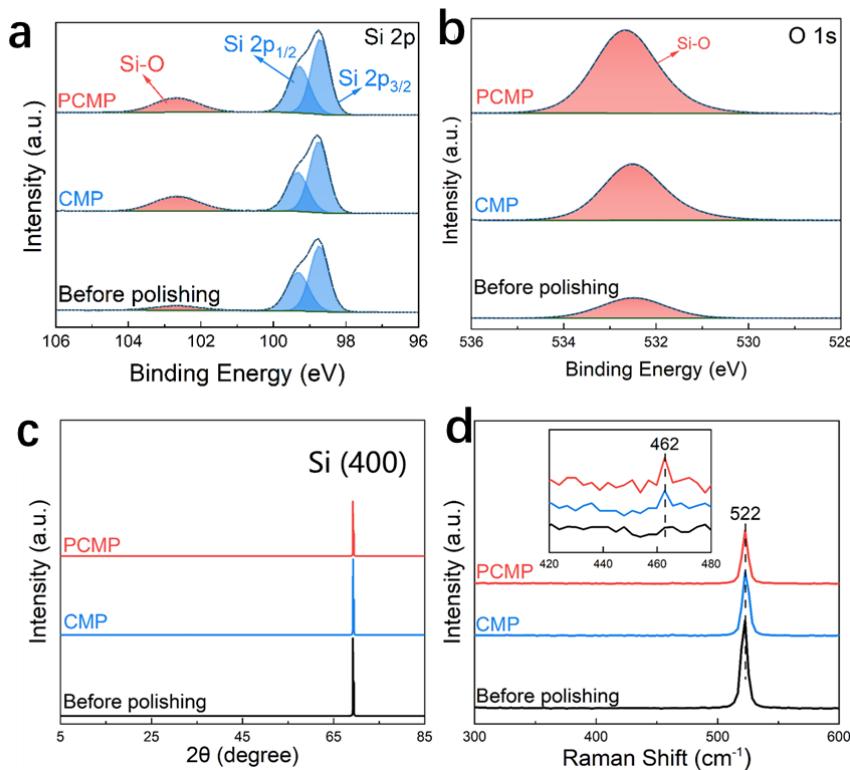
575 **Discussion:** AFM investigations visually supported the oxidation
 576 processes. The Sq of the initial Si wafer was measured at 0.174 nm, with
 577 a PV of 0.602 nm, indicating a relatively smooth Si surface with minimal
 578 evidence of corrosion or surface alteration (Fig. S39a). With the
 579 progressive introduction of CeO₂ particles, slurry components, and UV
 580 light, the Sq increased gradually, reaching a final value of 0.524 nm, and
 581 the peak-to-valley height reached 2.48 nm (Fig. S39b-d).



582 **Supplementary Fig. S40.** The SEM images, and EDS elemental mapping for Si and
 583 O are shown for **a** Si wafer before immersion, **b** CeO₂ particles, **c** CeO₂ slurry, and **d**
 584 CeO₂ slurry with UV illumination.

585

586 **Discussion:** SEM analyses further confirmed the formation of an
 587 oxidation layer (Fig. S40), with elemental mapping showing a significant
 588 increase in oxygen content (Table S5). Compared to the blank sample, the
 589 oxygen content (both in weight and atomic percentages) increases
 590 progressively with the addition of CeO₂ particles, CeO₂ slurry, and CeO₂
 591 slurry under UV irradiation. Specifically, the atomic percentage of
 592 oxygen increases from 3.31% (blank) to 14.69% (CeO₂ slurry + UV),
 593 indicating a substantial enhancement in surface oxidation.



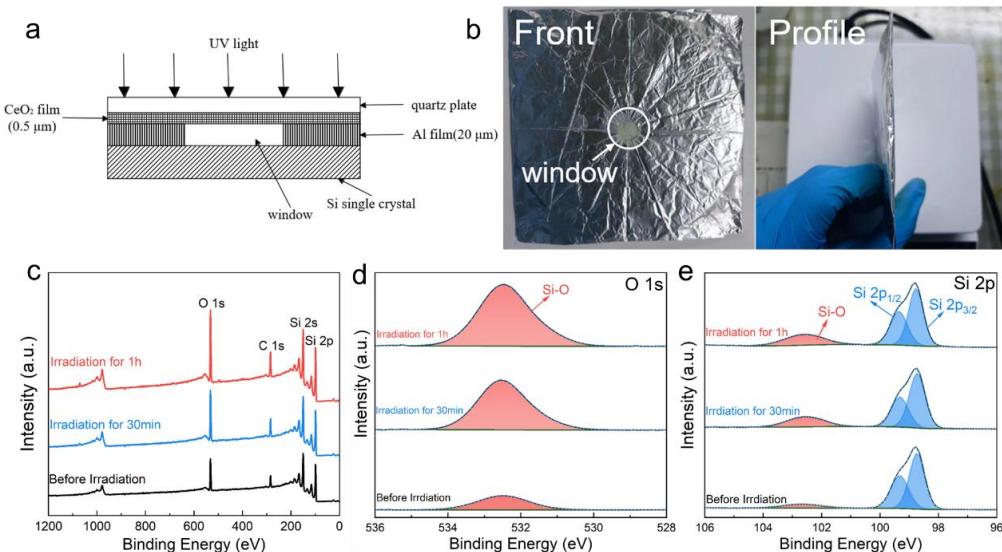
594 **Supplementary Fig. S41.** **a** Si 2p, **b** O 1s XPS spectra of Si wafers surface under
 595 different conditions. **c** XRD spectra. **d** Raman spectra of Si wafers surface under
 596 different conditions.

597

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

604 XRD patterns of the Si surface before polishing, after CMP, and
 605 after PCMP show a single diffraction peak at 69.63° corresponding to the
 606 Si (400) plane, with no additional peaks observed for SiO₂. This absence
 607 is attributed to the extremely thin and poorly crystalline nature of the
 608 surface oxide layer, making it undetectable by XRD (Fig. S41c). In
 609 contrast, Raman spectroscopy reveals a clear signal at 462 cm⁻¹ 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 SiO₂ layer to monocrystalline
614 silicon after CMP is 0.0237. In contrast, this ratio increases to 0.0363
615 after photocatalytic CMP, suggesting increased SiO₂ layer thickness.

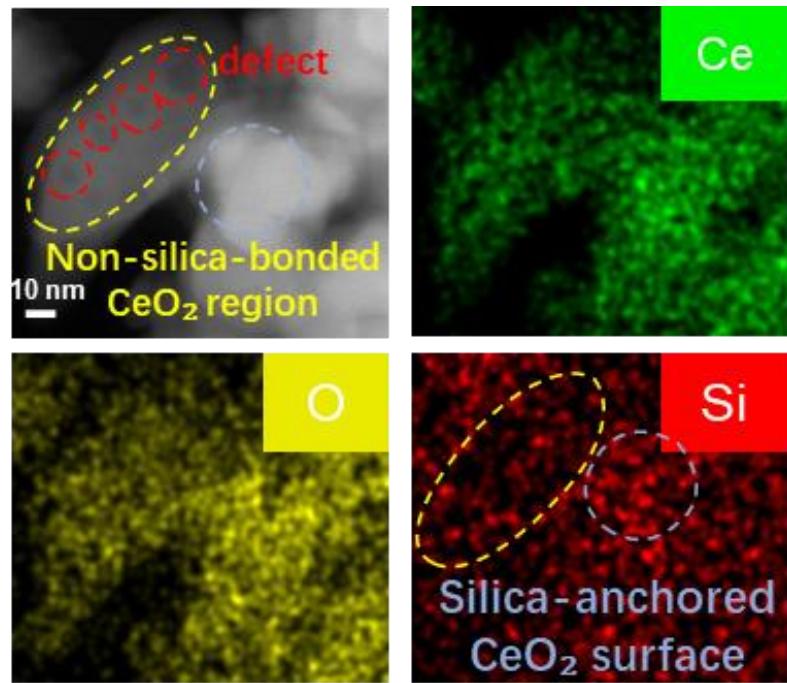


616 **Supplementary Fig. S42.** **a** Schematic illustration of the noncontact photocatalytic
617 oxidation system for the Si wafer surface. **b** Digital image of the noncontact
618 photocatalytic oxidation. **c** XPS spectra, **d** O 1s XPS spectra. **e** Si 2p XPS spectra of
619 Si wafer surface under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

620

621 **Discussion:** An aluminum metal film, approximately 20 μm thick, was
622 placed between the quartz plate coated with the CeO₂ film and the Si
623 substrate to maintain a gap distance of 20 μm between the CeO₂ film and
624 the Si surface, ensuring noncontact conditions¹⁵.

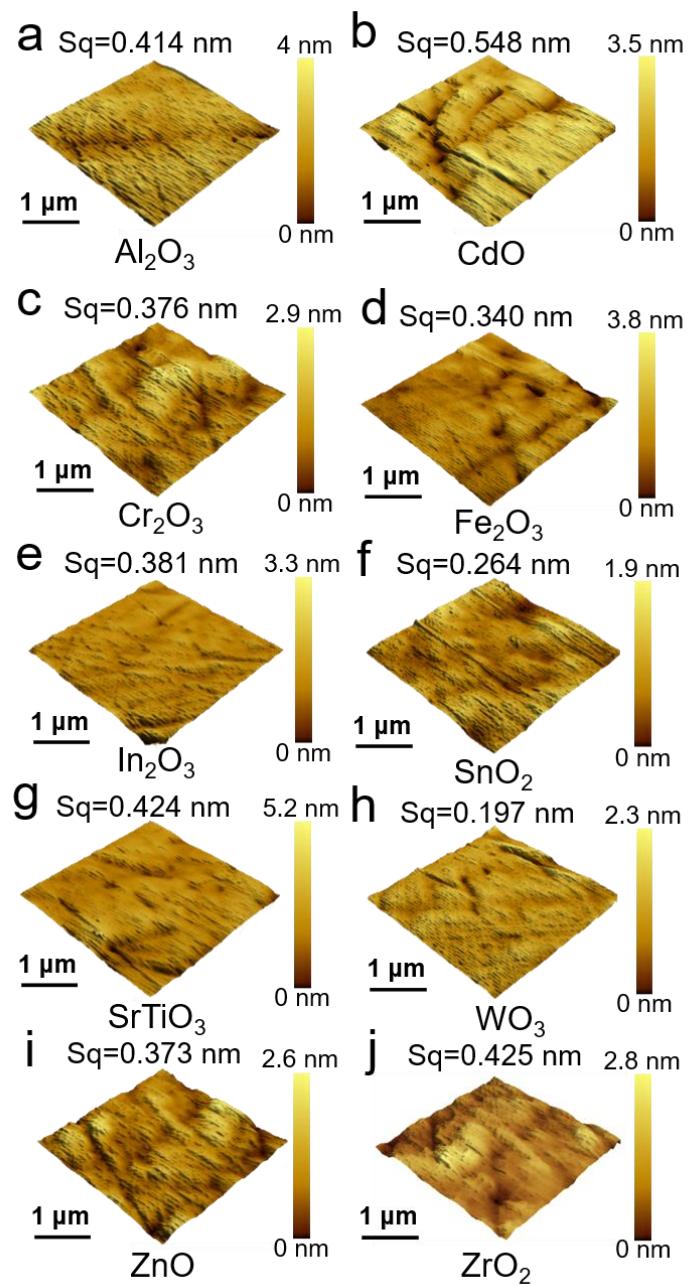
625 XPS spectra of the Si surface after irradiation for 0 h, 0.5 h, and 1 h
626 demonstrated that SiO₂ formation could occur on the Si surface via
627 photocatalytic oxidation even without direct contact between the CeO₂
628 film and the Si surface. The peaks at 532.40 and 532.20 eV binding
629 energies in the O 1s XPS spectrum correspond to SiO₂ and Si-O,
630 respectively. In the Si 2p XPS spectrum, the peaks at 98.70 and 99.30 eV
631 are attributed to Si, while the peaks at 103.30 and 102.40 eV correspond
632 to SiO₂ and Si-O, respectively. These results indicate that, even under
633 noncontact conditions, the Si surface undergoes oxidation to form SiO₂
634 via photocatalytic activity from CeO₂, with longer irradiation times
635 resulting in a more pronounced oxidation effect.



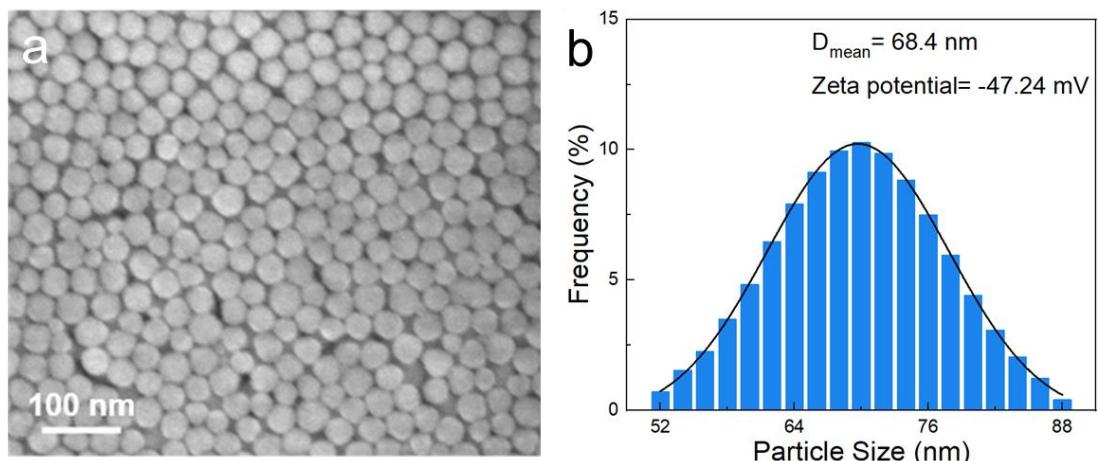
636 **Supplementary Fig. S43.** HAADF-STEM image and corresponding EDS elemental
 637 mappings of Ce (green), O (yellow), and Si (red) for CeO₂ particles after polishing a
 638 silicon wafer.

639

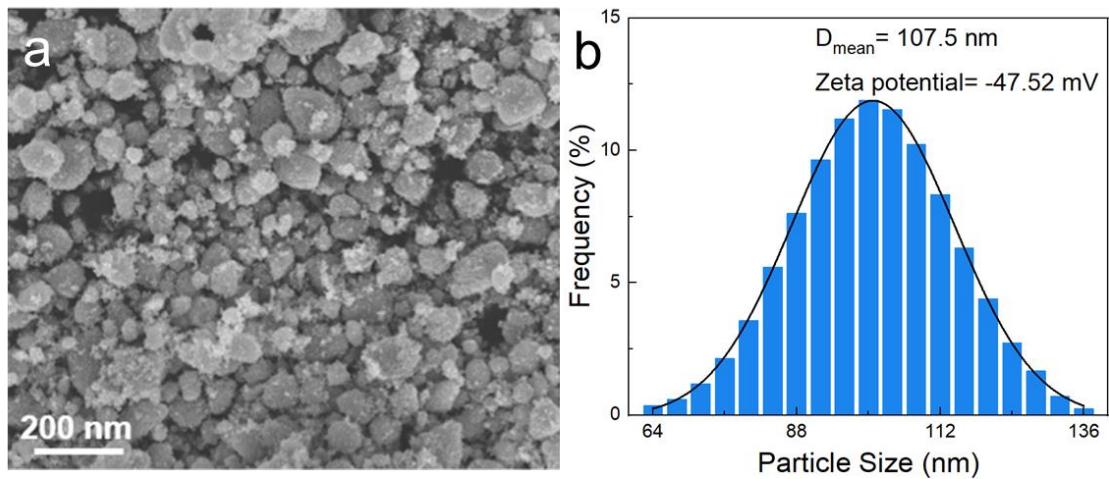
640 **Discussion:** To elucidate the interfacial interactions between CeO₂
 641 abrasives and the silicon wafer during PCMP, HAADF-STEM and EDS
 642 mapping were performed on CeO₂ after polishing. The yellow dashed
 643 region marks a CeO₂ domain not bonded to silica, characterized by
 644 apparent structural defects. In contrast, the blue dashed region shows a
 645 silica-anchored CeO₂ surface, where strong co-localization of Ce, O, and
 646 Si signals suggests the formation of interfacial Ce–O–Si bonding.



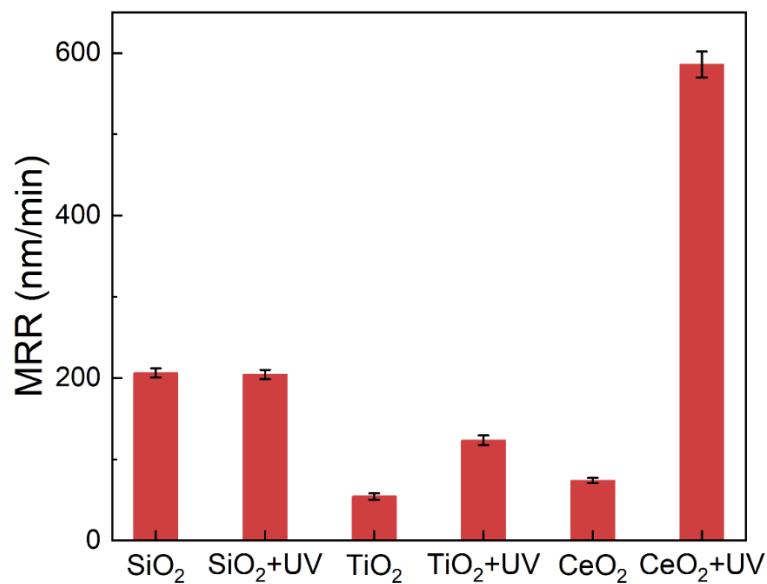
647 **Supplementary Fig. S44.** 3D AFM images of Si wafer surfaces polished under
 648 different abrasives: **a** Al_2O_3 ; **b** CdO ; **c** Cr_2O_3 ; **d** Fe_2O_3 ; **e** In_2O_3 ; **f** SnO_2 ; **g** SrTiO_3 ; **h**
 649 WO_3 ; **i** ZnO ; **j** ZrO_2 .



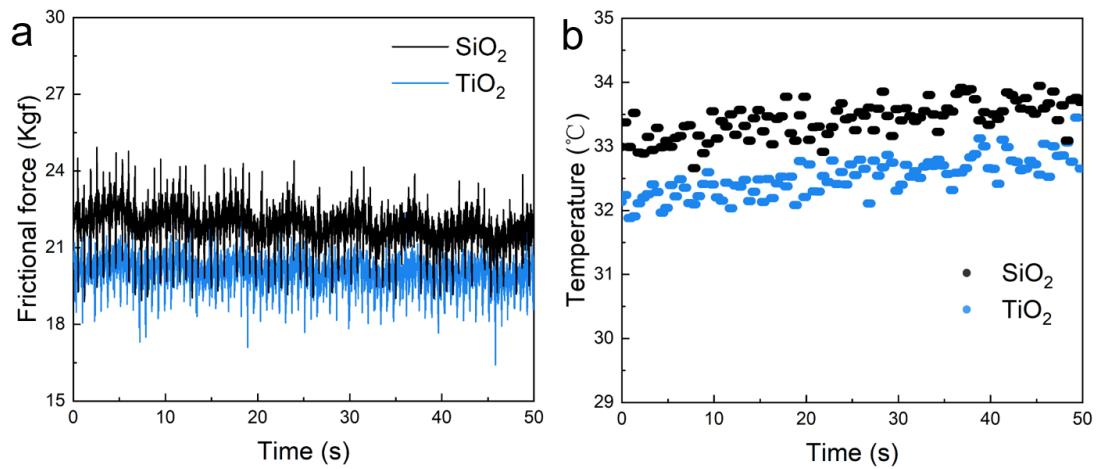
650 **Supplementary Fig. S45.** **a** SEM image of nano SiO_2 . **b** Particle size distribution
651 histogram and zeta potential of 2.5 wt.% SiO_2 exhibits an average SiO_2 particle size of
652 68.4 nm and a zeta potential of -47.24 mV.



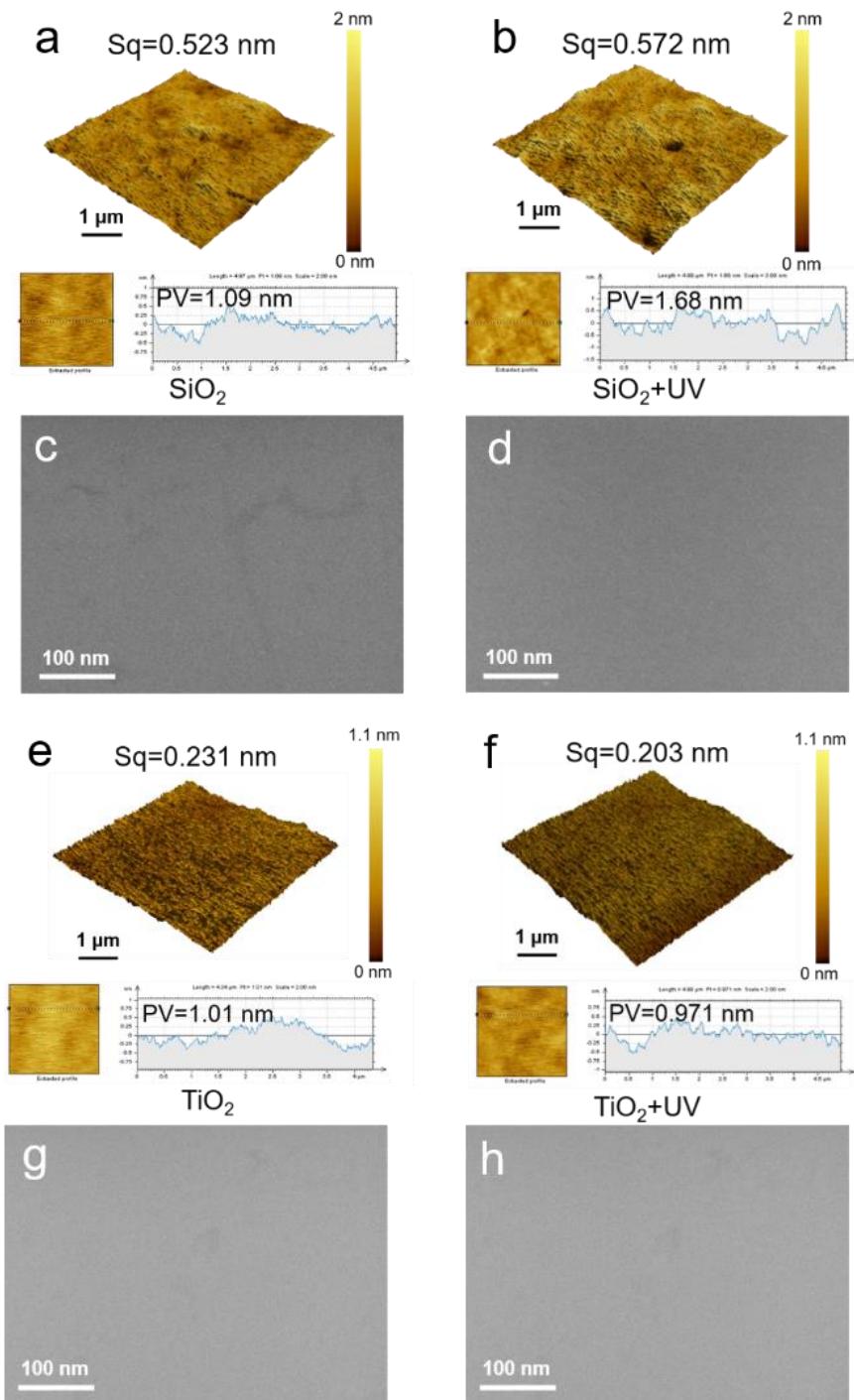
653 **Supplementary Fig. S46.** **a** SEM image of nano TiO₂. **b** Particle size distribution
654 histogram and zeta potential of 2.5 wt.% TiO₂ exhibits an average TiO₂ particle size
655 of 107.5 nm and a zeta potential of -47.52 mV.



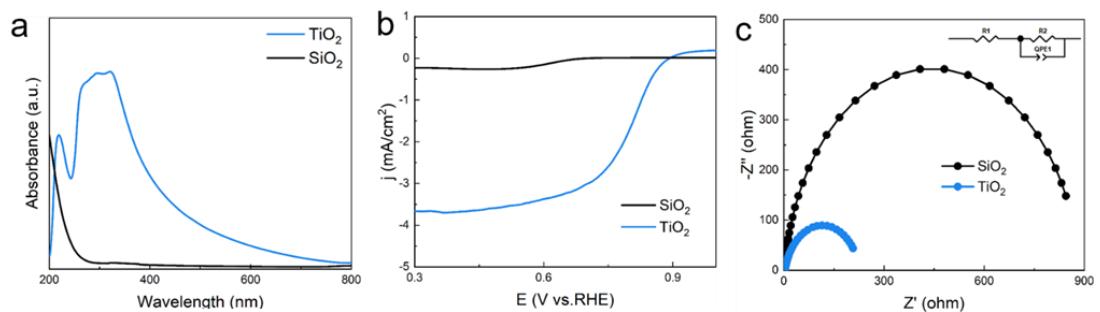
656 **Supplementary Fig. S47.** Comparison of MRR on the Si wafers by different slurries
657 (SiO_2 , TiO_2 , CeO_2) containing 2.5 wt% H_2O_2 under various conditions of UV light at
658 pH 10.



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_2 ; **b, d** 2.5 wt%
 663 SiO_2 ; **e, g** 2.5 wt% TiO_2 ; **f, h** 2.5 wt% TiO_2 with UV light irradiation.



664 **Supplementary Fig. S50.** **a** UV-Vis spectra. **b** Polarization curves. **c** Nyquist plot of
665 SiO_2 and TiO_2 .

666 **Supplementary Tables**

667

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

Sample	Ce ³⁺ (%)	Ce ⁴⁺ (%)	Ov (%)
CeO ₂ particles	21.34	78.66	30.17
CeO ₂ slurry	26.52	73.48	34.02

670

671 **Supplementary Table S2.** EXAFS fitting parameters at the Ce L₃-edge for various
 672 samples.

Sample	Shell	<i>CN</i> ^a	<i>R</i> (Å) ^b	σ (Å ²) ^c	ΔE_0 (eV) ^d	<i>R</i> factor
CeO ₂ reference	Ce-O	8	2.31	0.015	5.26	0.015
CeO ₂ particles	Ce-O	4.97	2.32	0.0091	6.32	0.019
CeO ₂ slurry	Ce-O	4.40	2.34	0.0103	6.82	0.014

673 ^a*CN*, coordination number; ^b*R*(Å), the distance to the neighboring atom; ^c σ (Å²),
 674 Debye-Waller factor to account for both thermal and structural disorders; ^d ΔE_0 inner
 675 potential correction; *R* factor indicates the goodness of the fit.

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

Component	E_{corr} vs. SCE (V)	I_{corr} (A/cm ²)
Blank	-0.222	6.561×10^{-7}
CeO ₂ particles	-0.257	1.143×10^{-6}
CeO ₂ slurry	-0.278	2.237×10^{-6}
CeO ₂ slurry+UV	-0.323	4.502×10^{-6}

677

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

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

680

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 ₂ particles	97.65	2.35	95.88	4.12
CeO ₂ slurry	94.22	5.78	90.35	9.65
CeO ₂ slurry+UV	91.01	8.99	85.31	14.69

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

	$\Delta H/\text{eV}$ (Total)	$\Delta H/\text{eV}$ (Per group)	$T \cdot \Delta S/\text{eV}$ (Per group)	$\Delta G/\text{eV}$ (Per group)
H_2O_2	-75.34	-4.70	-0.53	-4.17
$\cdot\text{OH}$	-26.76	-1.67	-0.72	-0.94
$\cdot\text{O}_2^-$	-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 (nm/min)	Surface roughness (nm)	Ret.
CeO ₂ /Na ₄ P ₂ O ₇ /H ₂ O ₂	90 rpm,70 rpm,30 Kpa, 15 mL/min,pH=10	127	0.067	¹⁶
SiO ₂ /H ₂ O ₂	40 rpm,40 rpm,7 psi, 7 L/min,alkaline	945	0.94	¹⁷
SiO ₂ /KNO ₃ /H ₂ O ₂	87 rpm,80 rpm,2.8 psi, 80 mL/min,pH=10.5	177.87	0.887	¹⁸
SiO ₂ /NH ⁴⁺ /H ₂ O ₂	87 rpm,80 rpm,2.5 psi, 80 mL/min,pH=10.5	168.7	0.744	¹⁹
amino-modified SiO ₂	80 rpm,80 rpm,2.64 psi, 90 g/min,pH=10.6	132.53	0.219	²⁰
SiO ₂ /EDA/H ₂ O ₂	71 rpm,69 rpm,5.7 psi, 100 mL/min,pH=10.9	552.8	0.353	²¹
SiO ₂ /C ₆ H ₅ O ₇ (NH ₄) ₃ /H ₂ O ₂	87 rpm,80 rpm,2.8 psi, 80 mL/min,pH=10.5	197.98	0.563	²²
CeO ₂ @ZIF-8/PVP	87 rpm,80 rpm,80 N, 100 mL/min	151.07	0.231	²³
SiO ₂ /H ₂ O ₂	180 rpm,16.7 KPa,10 mL/min,pH=9	60	0.4	²⁴
CeO ₂ /PhPs	180 rpm,16.7 KPa,10 mL/min,pH=9, UV irradiation(365 nm)	23	0.19	²⁴
SiO ₂ /MEA/H ₂ O ₂	40 rpm,40 rpm,2 psi,155 mL/min,pH=10.5	24	0.178	²⁵

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

689 **Supplementary Table S8.** The average friction force and temperature of Si wafers
690 polished with SiO₂, CeO₂, TiO₂ and CeO₂ slurry.

Abrasive	Friction force (Kgf)	Temperature (°C)
SiO ₂	23.33	32.62
CeO ₂ particles	21.65	33.08
TiO ₂	20.08	33.49
CeO ₂ slurry	21.63	32.97

691

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

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

694

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