

Supplementary Information for

Selective hydroxylation of carbon surfaces for long-lasting hydrophilicity by a green chemistry process

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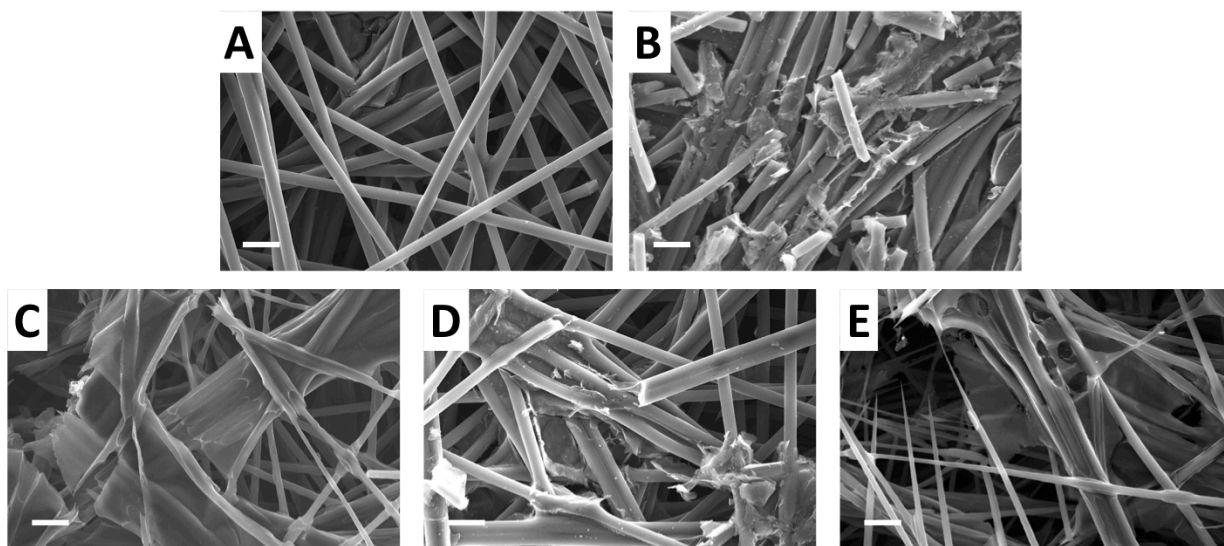


Fig. S1 SEM images of untreated and thermally annealed CFP. **A** Untreated CFP. **B** Heat-treated CFP according to a preparation by Zhang et al. *J Mater Chem A* **4**, 18639-18645 (2016). **C** Untreated CFP that was thermally annealed at 500 °C for 5 min. **D** Pretreated CFP that was thermally annealed at 500 °C for 5 min. **E** Pretreated CFP that was thermally annealed at 800 °C for 5 min. Pretreatment consisted of immersion in piranha solution, sonication in SDS solution, followed by electrooxidation. Clearly, all thermal treatments led to significant structural damage to the carbon fibers. All scalebars are 20 μm .

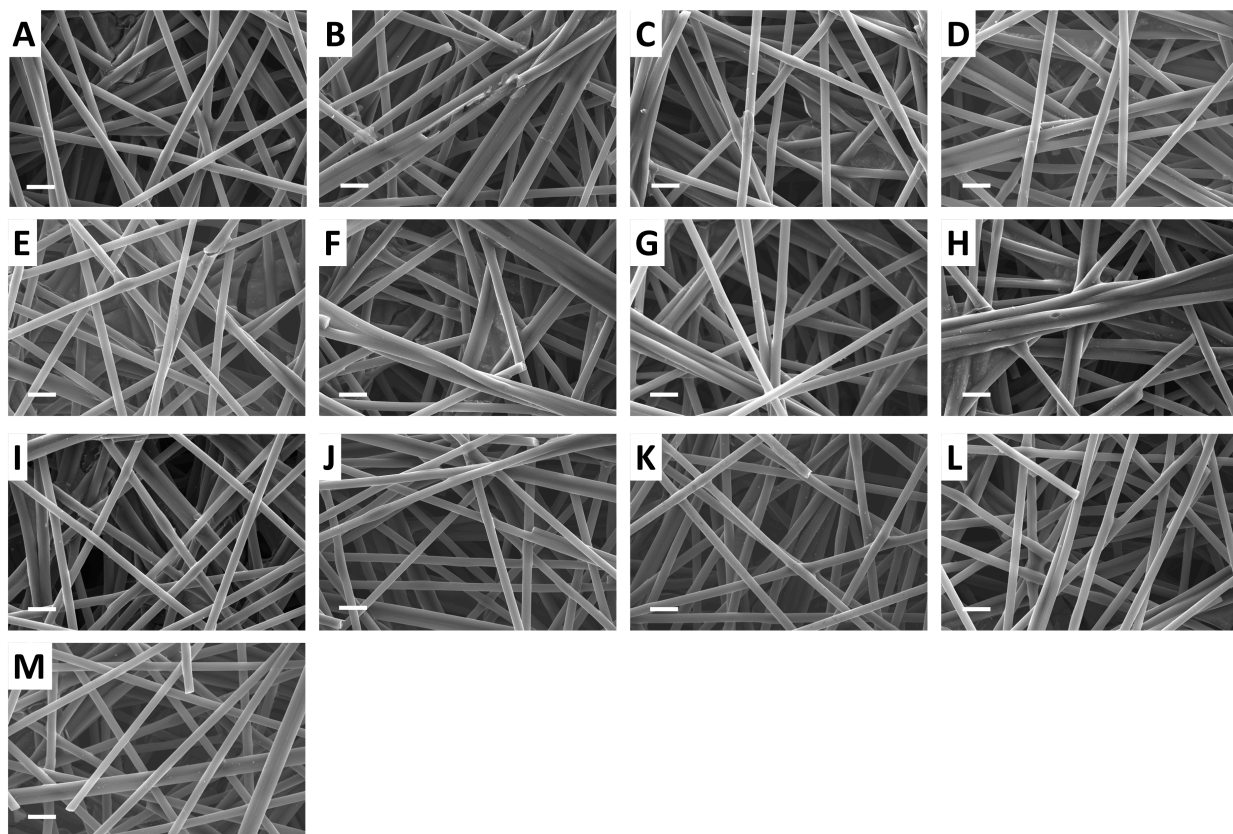


Fig. S2 SEM images of untreated and chemically treated CFP. Treatments of CFP: **A** none. **B** Piranha solution. **C** SDS solution. **D** Piranha and KOH solutions. **E** Piranha, SDS, and KOH solutions. **F** Piranha solution and concentrated formic acid. **G** Piranha, SDS solutions, and concentrated formic acid. **H** Piranha and SDS solutions. **I** Electrooxidation. **J** Piranha and SDS solutions plus electrooxidation. **K** Piranha solution plus electrooxidation. **L** Piranha solution and concentrated formic acid plus electrooxidation. **M** Piranha and KOH solutions plus electrooxidation. All scalebars are 20 μm .

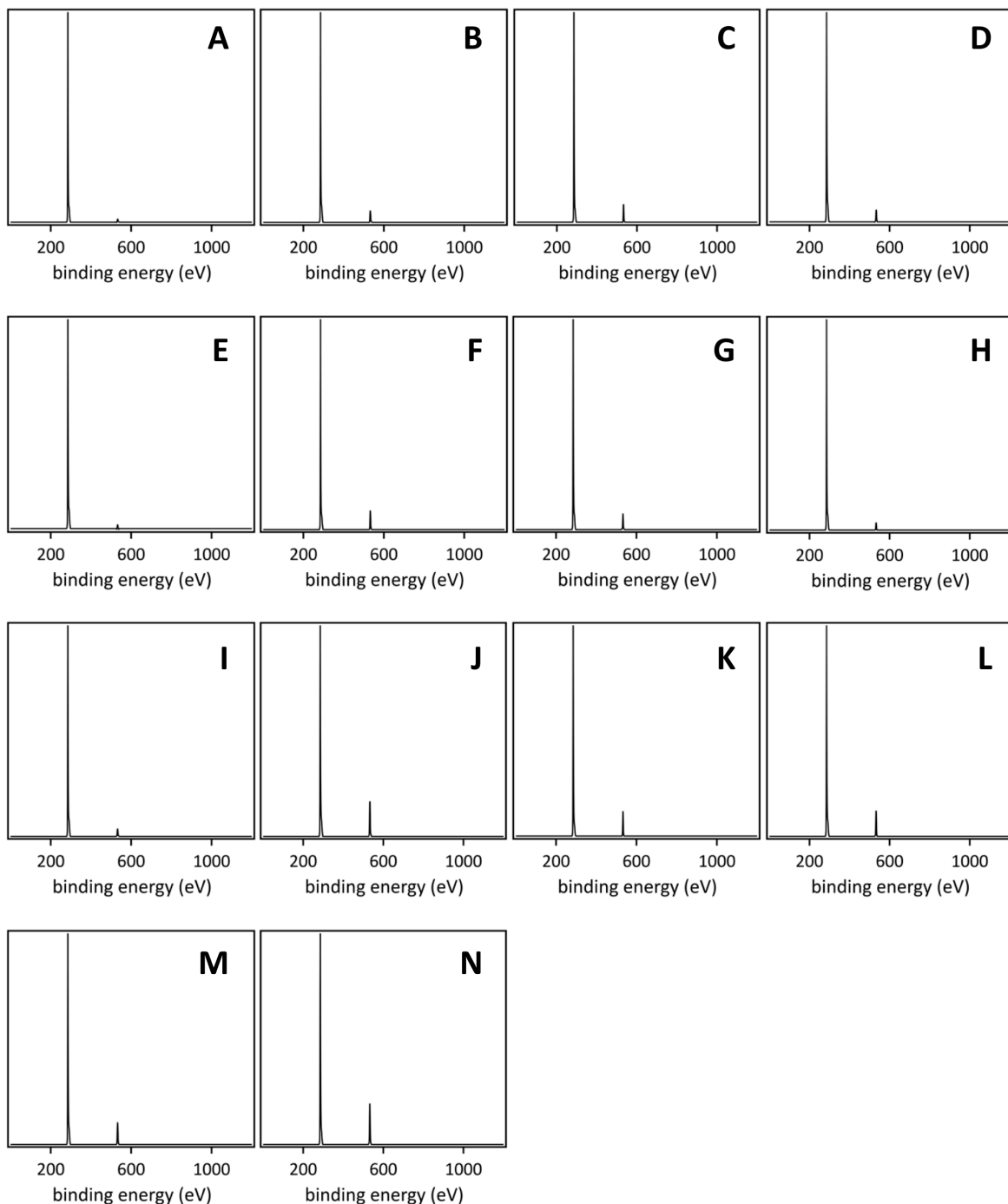


Fig. S3 Survey XPS data of untreated and chemically treated CFP. Treatments of CFP: **A** none. **B** Piranha solution. **C** SDS solution. **D** Piranha and KOH solutions. **E** Piranha, SDS, and KOH solutions. **F** Piranha solution and concentrated formic acid. **G** Piranha, SDS solutions, and concentrated formic acid. **H** Piranha and SDS solutions. **I** Electrooxidation. **J** Piranha and SDS solutions plus electrooxidation. **K** Piranha solution plus electrooxidation. **L** Piranha solution and concentrated formic acid plus electrooxidation. **M** Piranha and KOH solutions plus electrooxidation. **N** SDS solution plus electrooxidation.

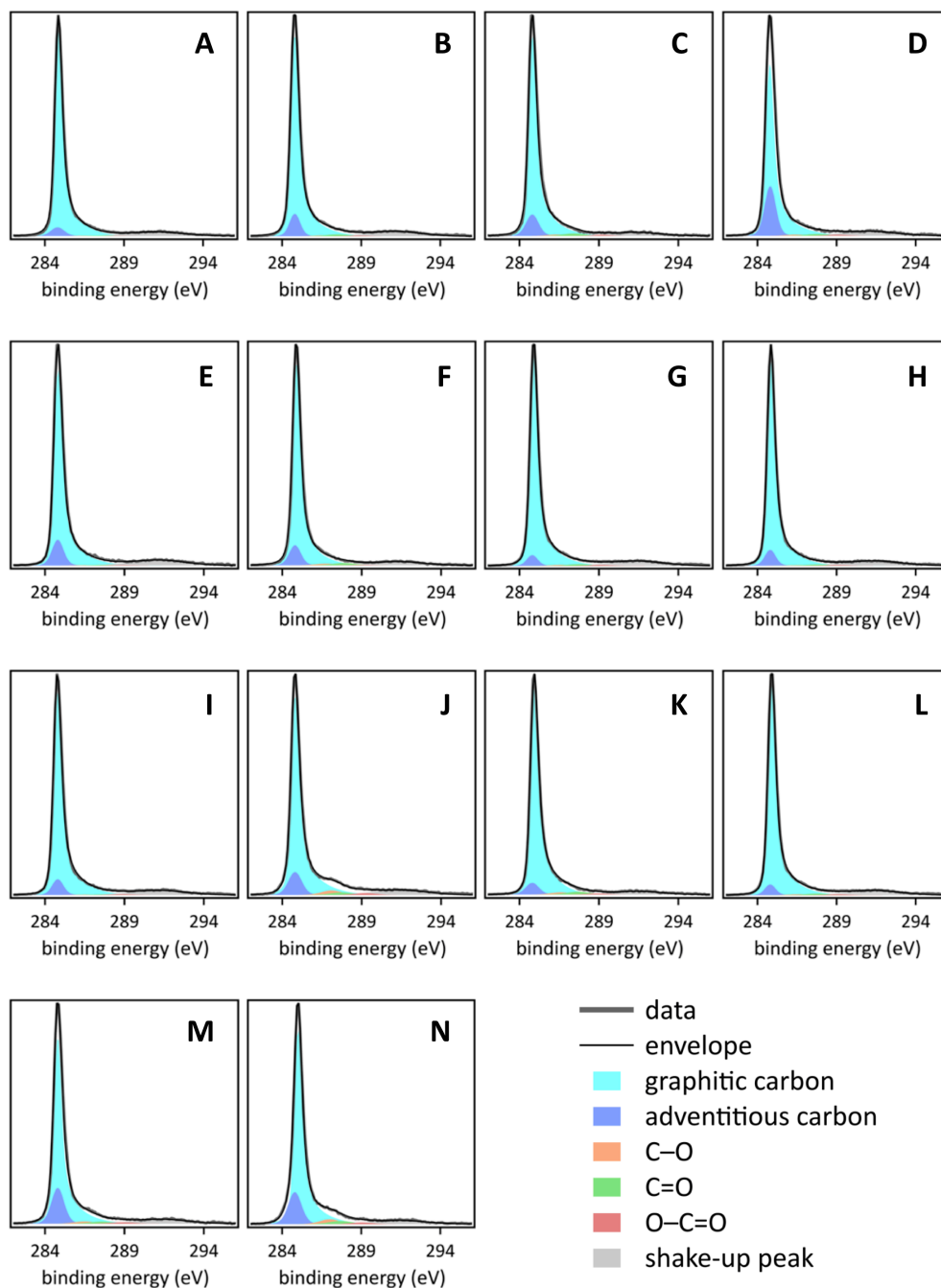


Fig. S4 High-resolution C 1s XPS data of untreated and chemically treated CFP. Treatments of CFP: **A** none. **B** Piranha solution. **C** SDS solution. **D** Piranha and KOH solutions. **E** Piranha, SDS, and KOH solutions. **F** Piranha solution and concentrated formic acid. **G** Piranha, SDS solutions, and concentrated formic acid. **H** Piranha and SDS solutions. **I** Electrooxidation. **J** Piranha and SDS solutions plus electrooxidation. **K** Piranha solution plus electrooxidation. **L** Piranha solution and concentrated formic acid plus electrooxidation. **M** Piranha and KOH solutions plus electrooxidation. **N** SDS solution plus electrooxidation.

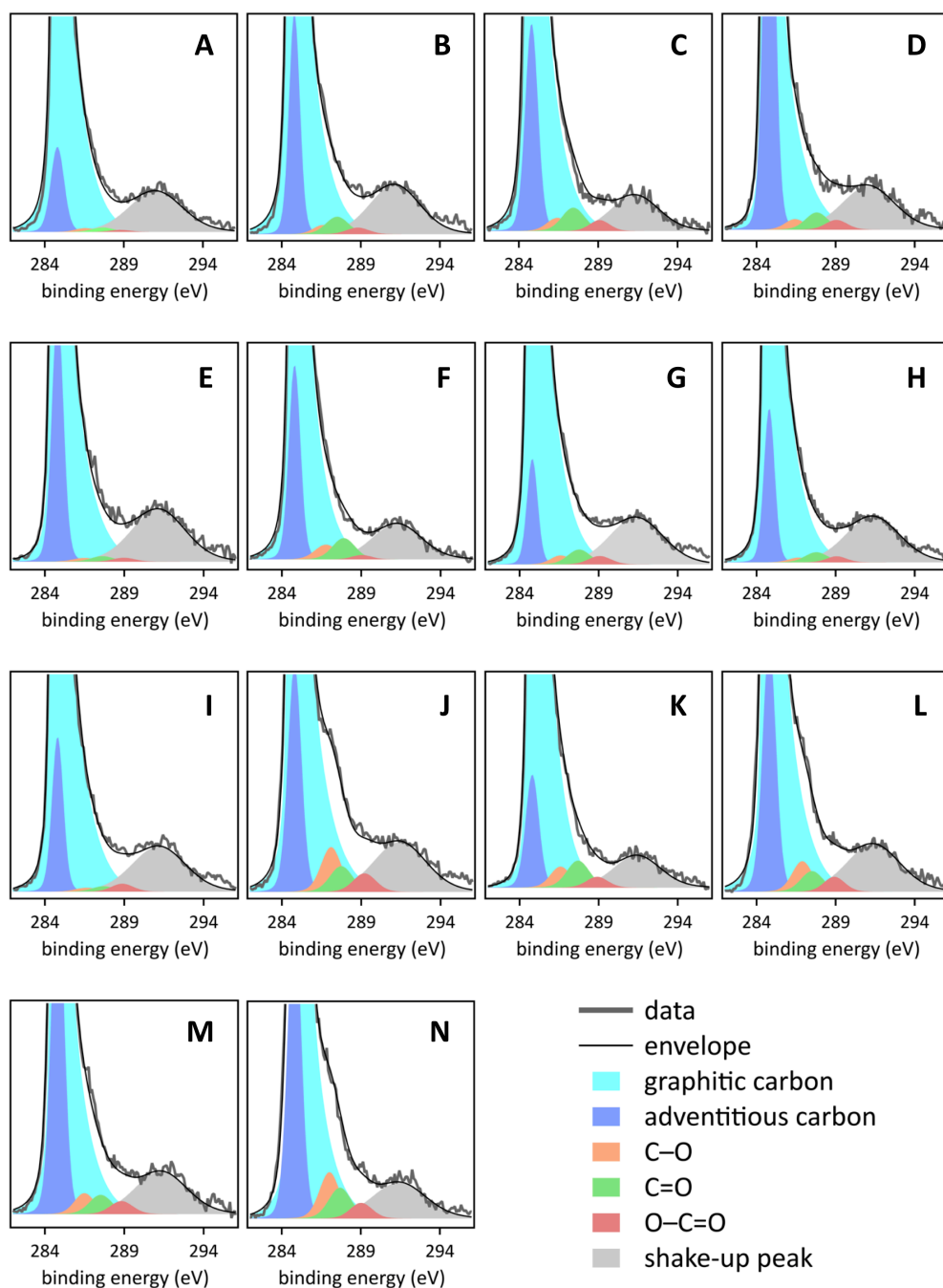


Fig. S5 High-resolution C 1s XPS data, with a y-axis that was magnified by a factor of 10, of untreated and chemically treated CFP. Treatments of CFP: **A** none. **B** Piranha solution. **C** SDS solution. **D** Piranha and KOH solutions. **E** Piranha, SDS, and KOH solutions. **F** Piranha solution and concentrated formic acid. **G** Piranha, SDS solutions, and concentrated formic acid. **H** Piranha and SDS solutions. **I** Electrooxidation. **J** Piranha and SDS solutions plus electrooxidation. **K** Piranha solution plus electrooxidation. **L** Piranha solution and concentrated formic acid plus electrooxidation. **M** Piranha and KOH solutions plus electrooxidation. **N** SDS solution plus electrooxidation.

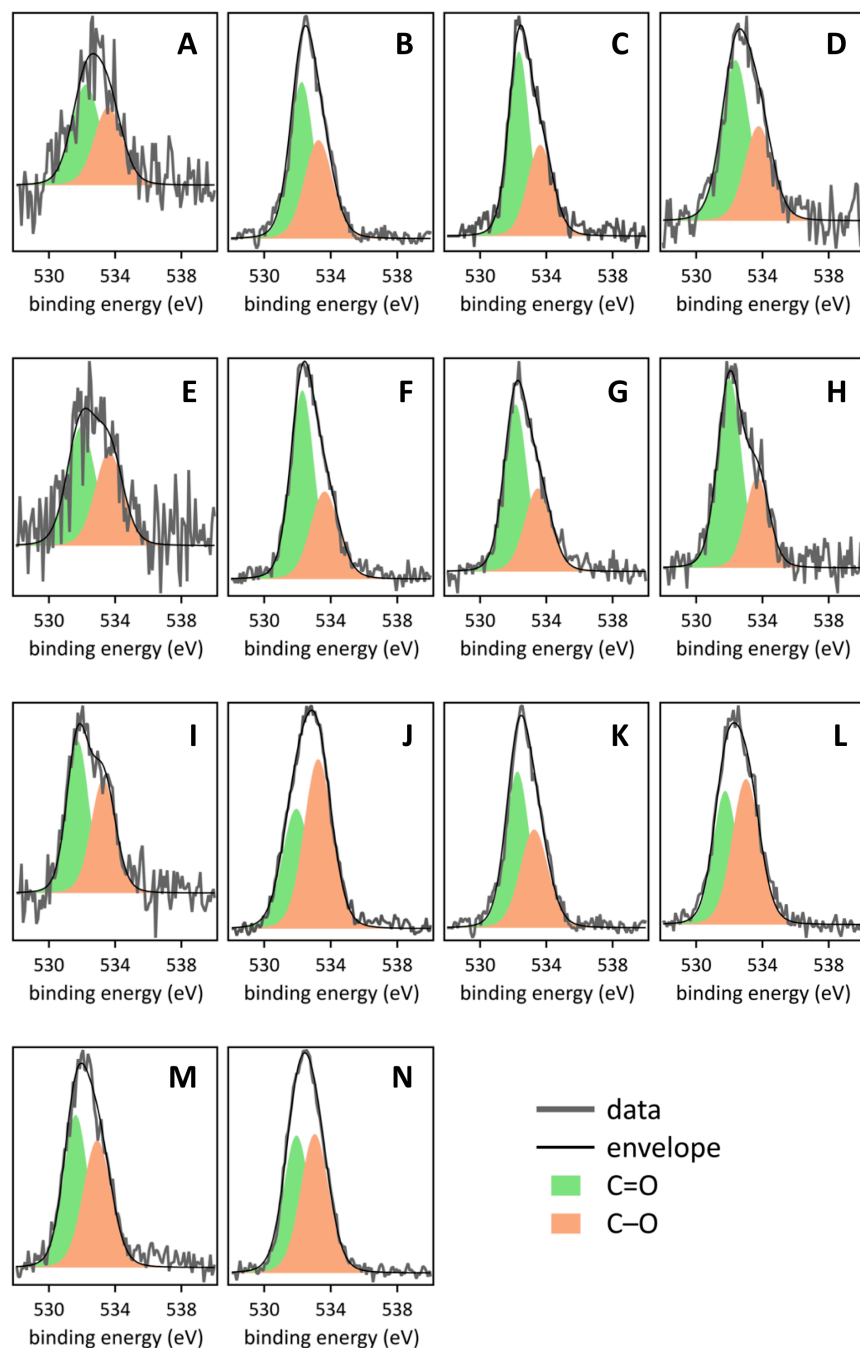


Fig. S6 High-resolution O 1s XPS data of untreated and chemically treated CFP. Treatments of CFP: **A** none. **B** Piranha solution. **C** SDS solution. **D** Piranha and KOH solutions. **E** Piranha, SDS, and KOH solutions. **F** Piranha solution and concentrated formic acid. **G** Piranha, SDS solutions, and concentrated formic acid. **H** Piranha and SDS solutions. **I** Electrooxidation. **J** Piranha and SDS solutions plus electrooxidation. **K** Piranha solution plus electrooxidation. **L** Piranha solution and concentrated formic acid plus electrooxidation. **M** Piranha and KOH solutions plus electrooxidation. **N** SDS solution plus electrooxidation.

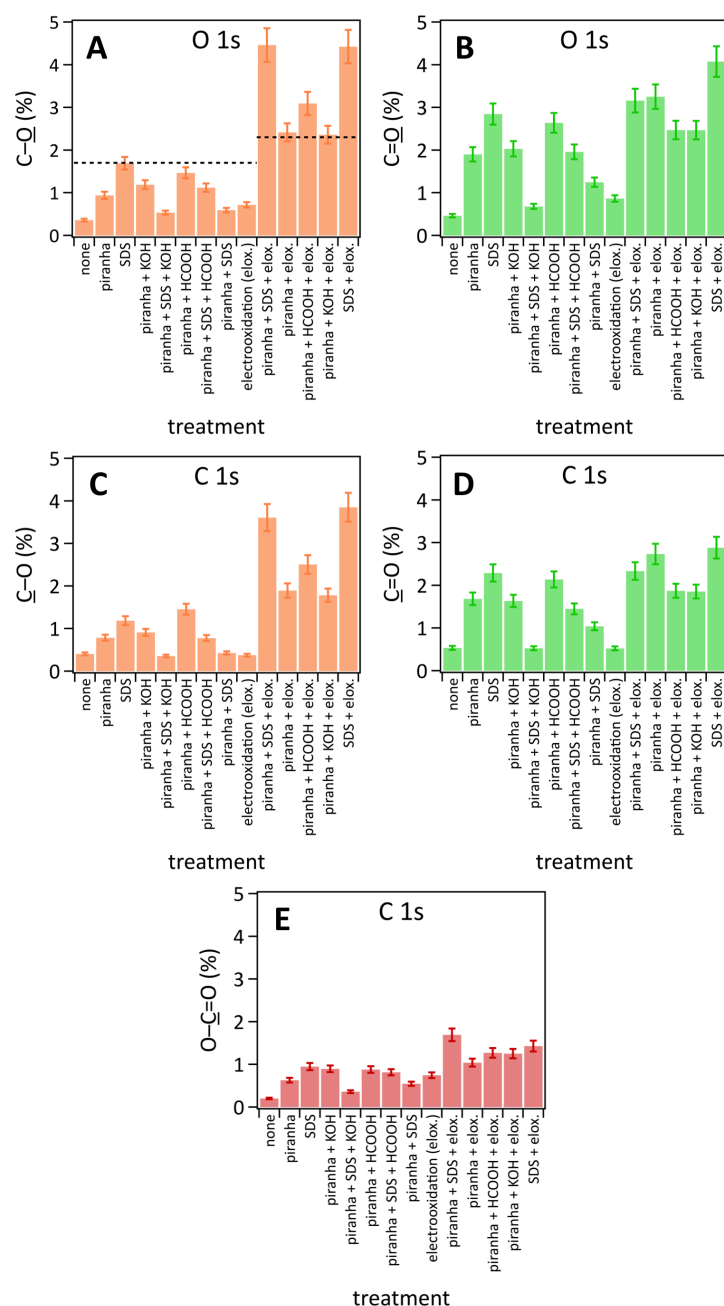


Fig. S7 Quantification of oxygenated carbon surface species. **A** $\text{C}-\text{Q}$; the dashed lines indicate surface $\text{C}-\text{Q}$ contents of 1.7% (left) and 2.3% (right). **B** $\text{C}=\text{Q}$. **C** $\text{C}-\text{O}$. **D** $\text{C}=\text{O}$. **E** $\text{O}-\text{C}=\text{O}$. Piranha refers to soaking for 5 minutes in a 4:1 (v/v) mixture of concentrated sulfuric acid and 30% aqueous hydrogen peroxide solution, SDS refers to sonication for 5 minutes in a 1 M aqueous sodium dodecyl sulfate solution, KOH refers to soaking for 5 minutes in a 6 M aqueous KOH solution, HCOOH refers to soaking for 5 minutes in concentrated formic acid, and electrooxidation (elox.) was performed in 0.1 M pH 8.7 aqueous KHCO_3 electrolyte at +1.63 V vs. Ag/AgCl for 20 minutes.

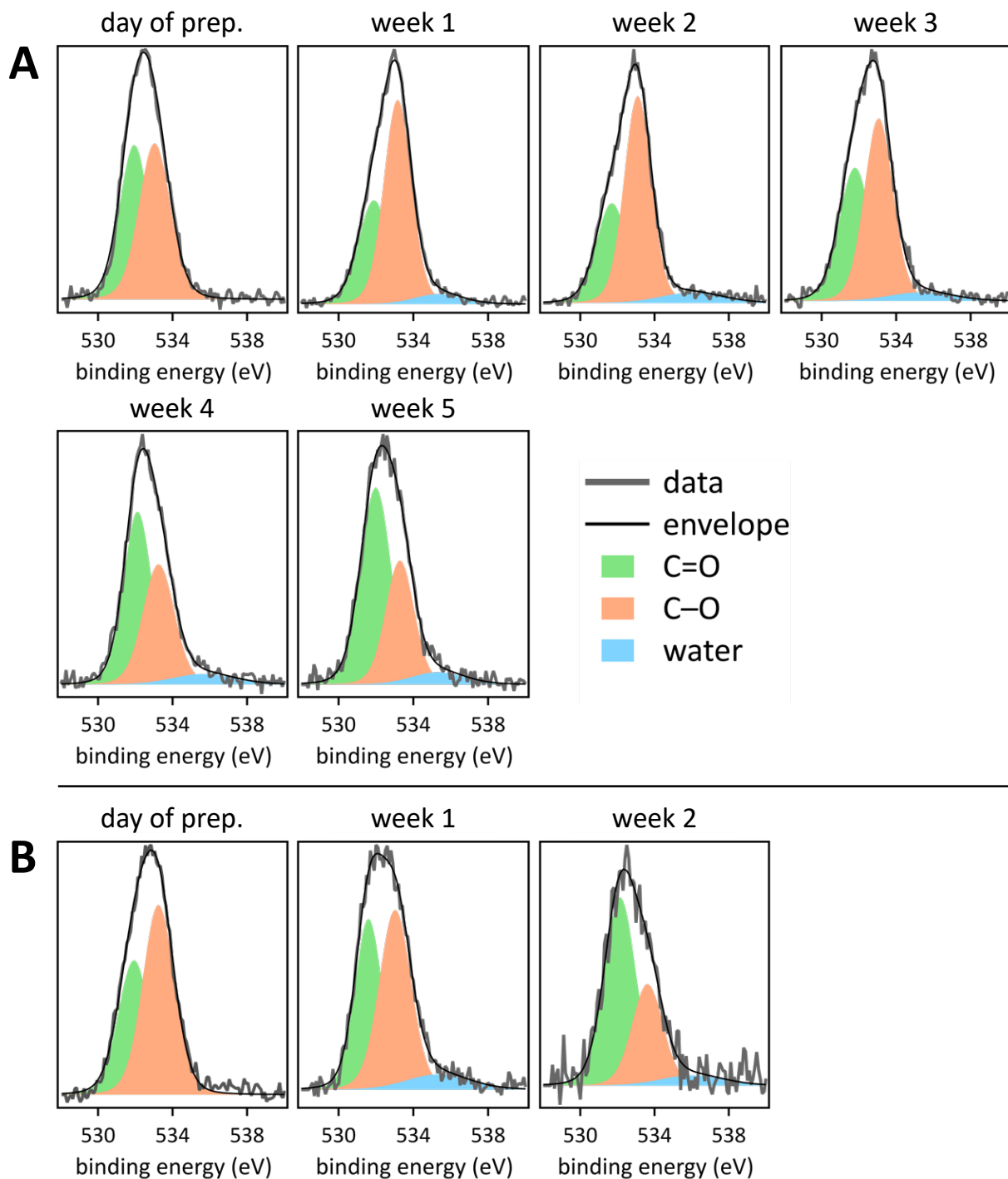


Fig. S8 Time evolution of O 1s signals. A SDS-electrooxidation treated CFP. **B** Piranha-SDS-electrooxidation treated CFP. Day of prep. means XPS data collected on the day of sample preparation. CFP was stored in ambient air.

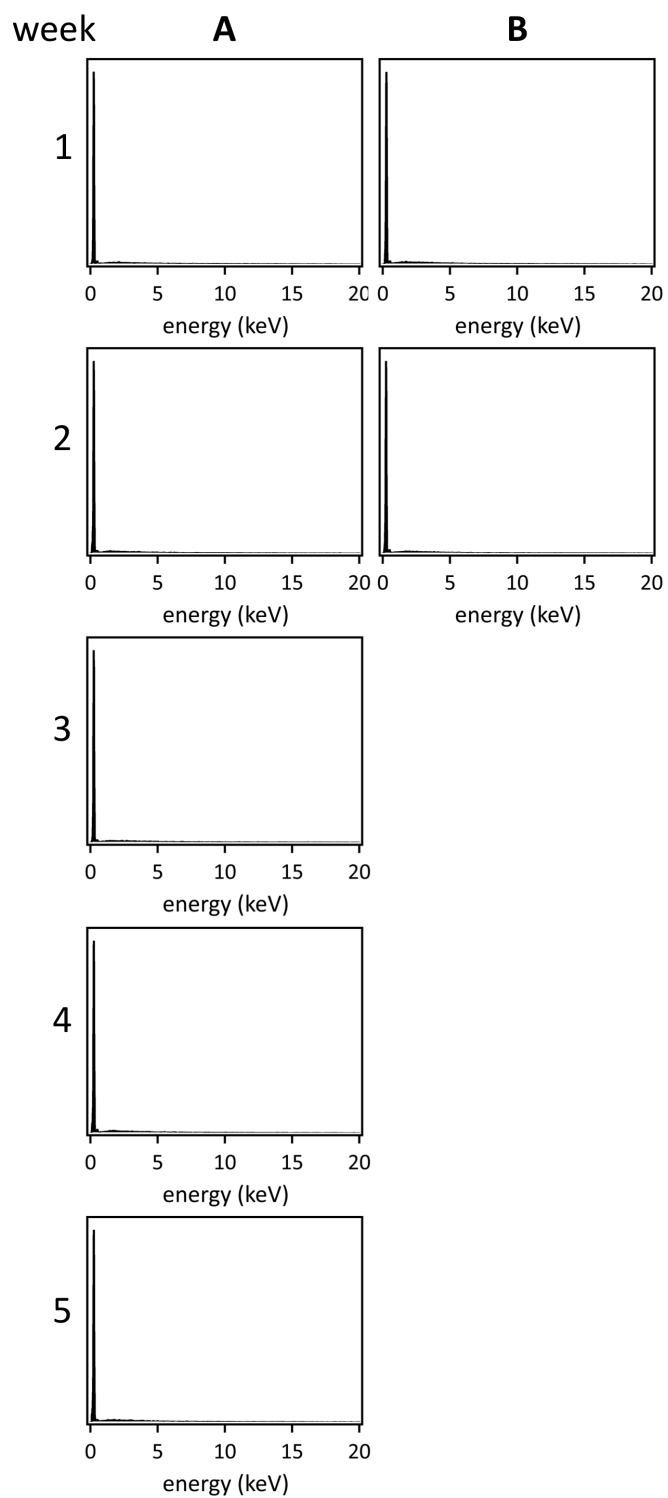


Fig. S9 Time evolution of CFP surface elements. SEM-EDX data of CFP that was stored in ambient air after treatment with **A** SDS solution plus electrooxidation, or **B** Piranha and SDS solutions plus electrooxidation.

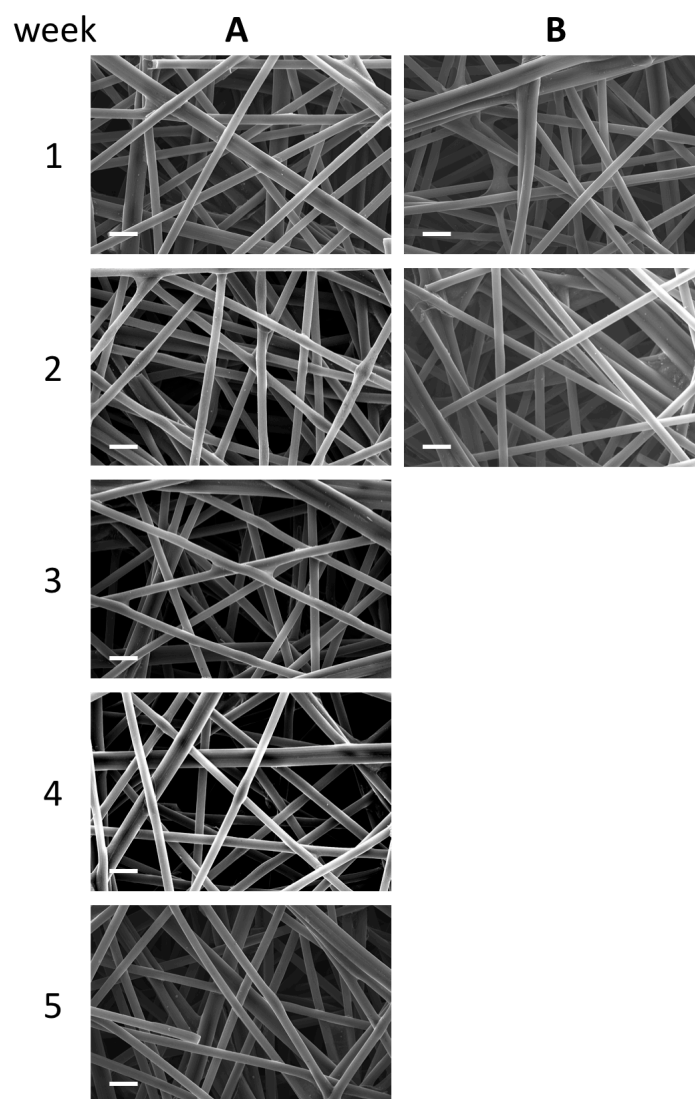


Fig. S10 Time evolution of CFP structures. SEM images of CFP that was stored in ambient air after treatment with **A** SDS solution plus electrooxidation, or **B** Piranha and SDS solutions plus electrooxidation. All scalebars are 20 μm .

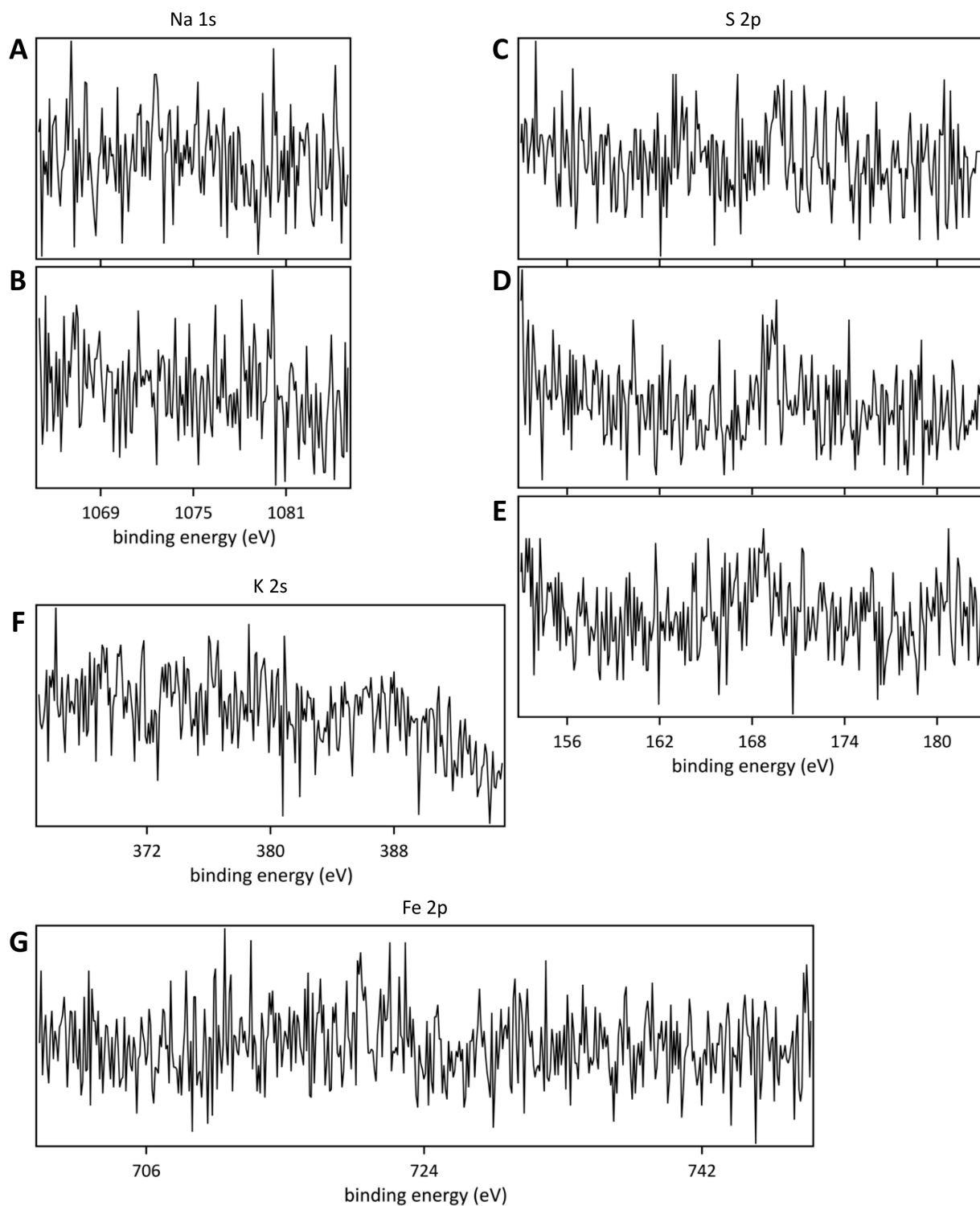


Fig. S11 XPS data of CFP indicating the absence of surface residues from chemical treatments. **A** Untreated CFP (Na 1s). **B** SDS–electrooxidation treated CFP (Na 1s). **C** Untreated CFP (S 2p). **D** SDS–electrooxidation treated CFP (S 2p). **E** Piranha–SDS–electrooxidation treated CFP (S 2p). **F** SDS–electrooxidation treated CFP (K 2s). **G** SDS–electrooxidation treated CFP (Fe 2p).

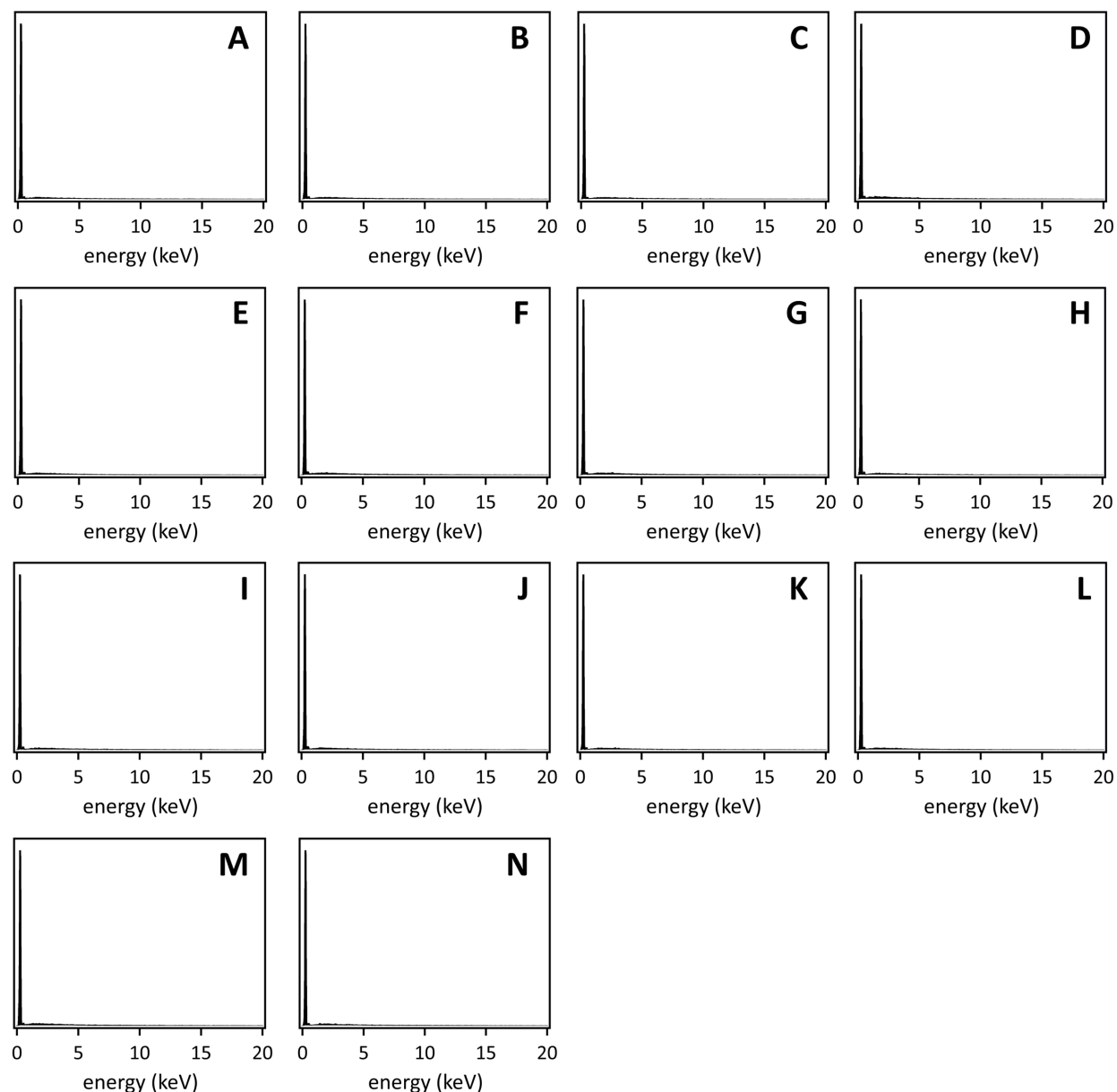


Fig. S12 SEM-EDX data of untreated and chemically treated CFP. Treatments of CFP: **A** none. **B** Piranha solution. **C** SDS solution. **D** Piranha and KOH solutions. **E** Piranha, SDS, and KOH solutions. **F** Piranha solution and concentrated formic acid. **G** Piranha, SDS solutions, and concentrated formic acid. **H** Piranha and SDS solutions. **I** Electrooxidation. **J** Piranha and SDS solutions plus electrooxidation. **K** Piranha solution plus electrooxidation. **L** Piranha solution and concentrated formic acid plus electrooxidation. **M** Piranha and KOH solutions plus electrooxidation. **N** SDS solution plus electrooxidation.

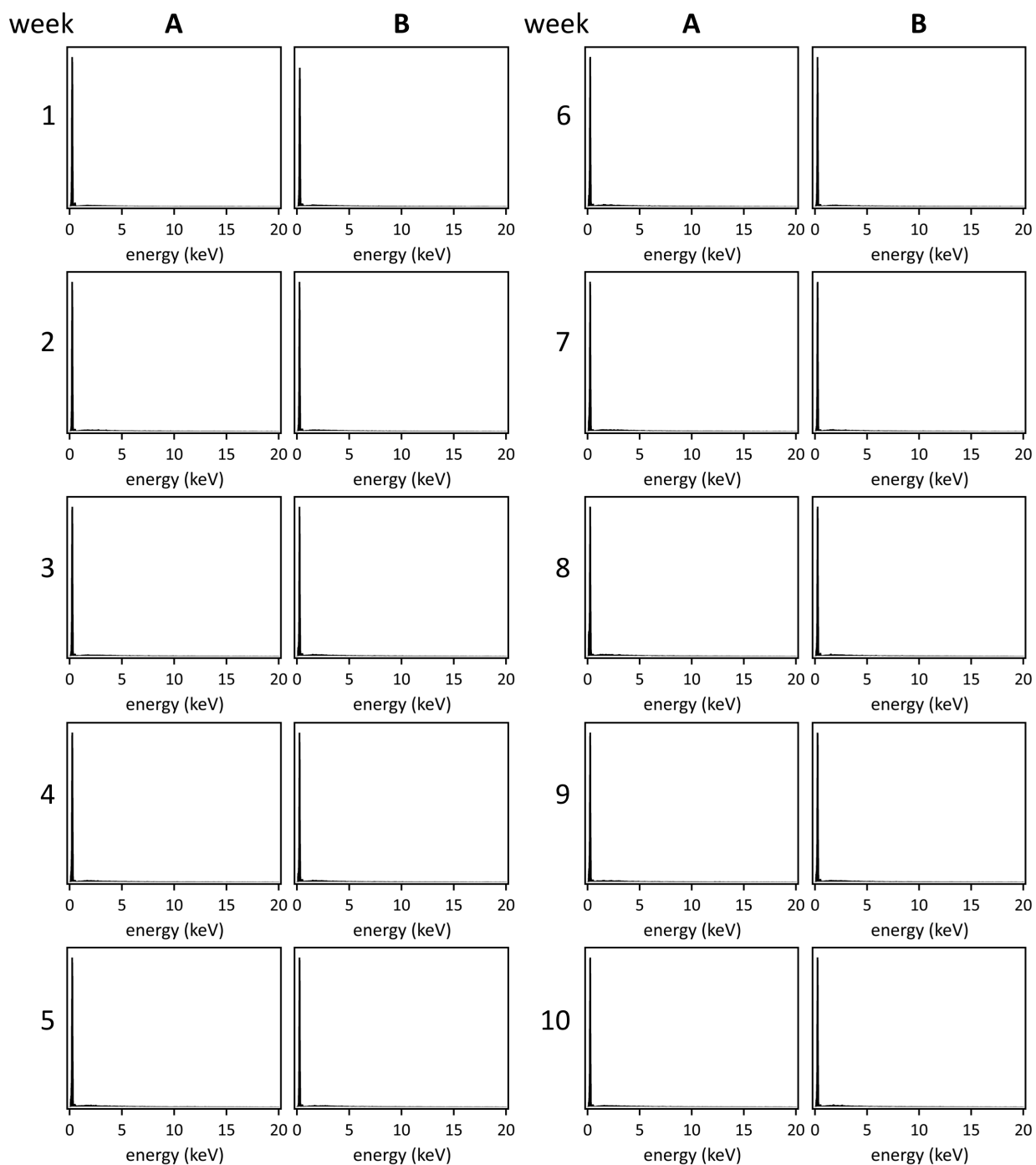


Fig. S13 Time evolution of CFP surface elements. SEM-EDX data of CFP that was stored under water after treatment with **A** SDS solution plus electrooxidation, or **B** Piranha and SDS solutions plus electrooxidation.

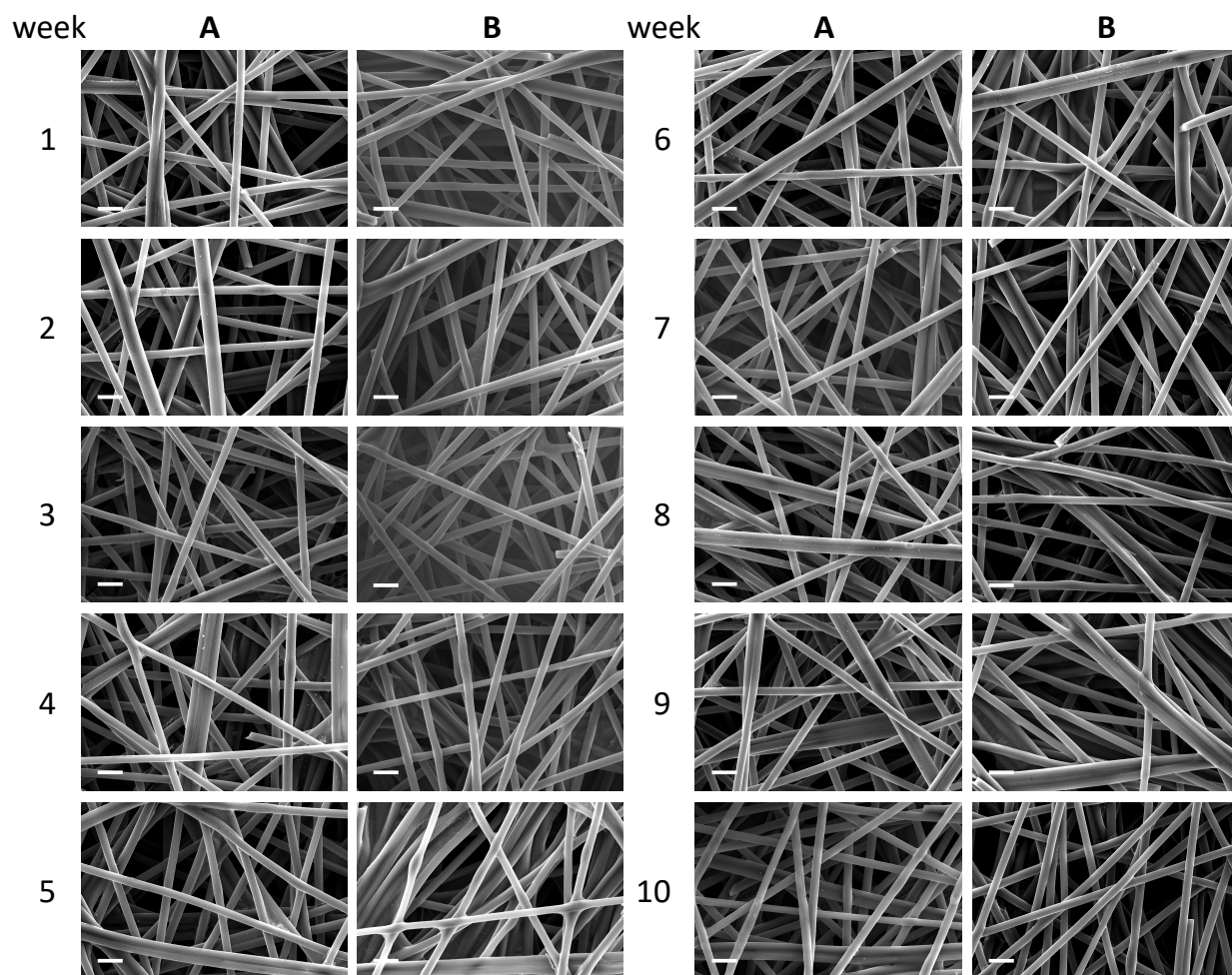


Fig. S14 Time evolution of CFP structures. SEM images of CFP that was stored under water after treatment with **A** SDS solution plus electrooxidation, or **B** Piranha and SDS solutions plus electrooxidation. All scalebars are 20 μm .

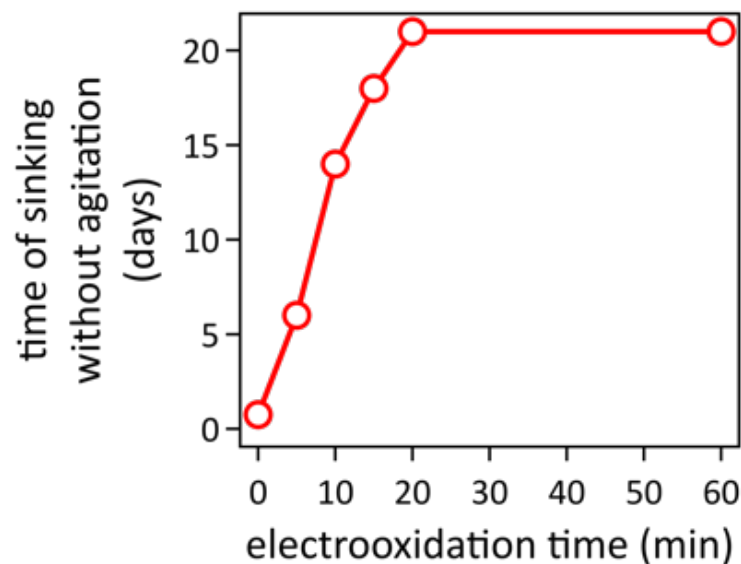


Fig. S15 Optimization of electrooxidation time. Time of sinking without agitation vs. electrooxidation time when CFP was stored in ambient air; circles, data points, lines guide the eye. CFP was treated by sonication in SDS solution, followed by electrooxidation in 0.1 M pH 8.7 aqueous KHCO_3 electrolyte at +1.63 V vs. Ag/AgCl for various times.

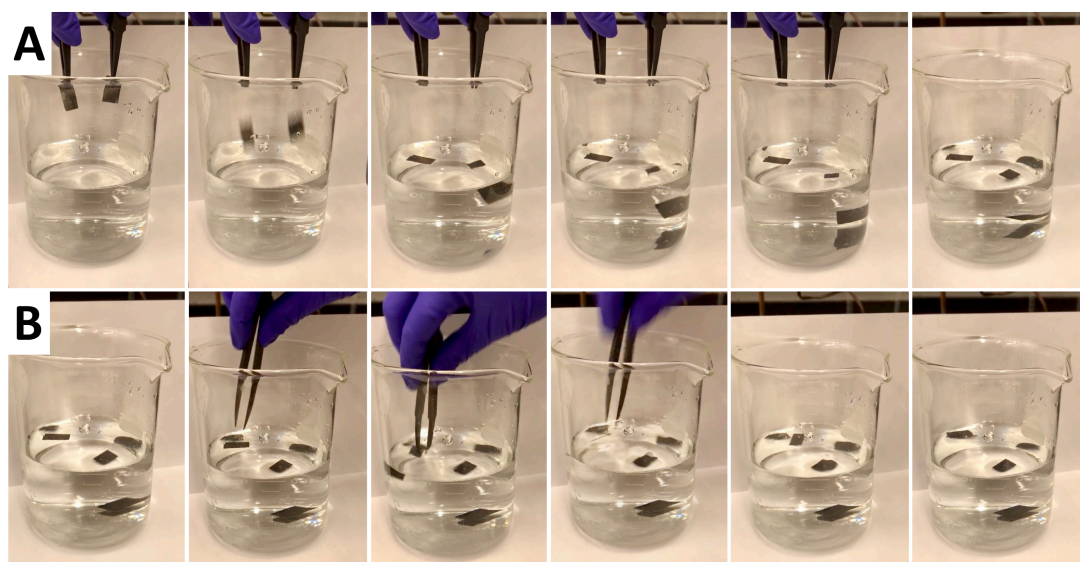


Fig. S16 Visualization of the sink-or-float test to monitor hydrophilicity of CFP. Photographs of sink-or-float testing of dry untreated (left) and dry treated (right) CFP pieces in deionized water. **A** Untreated CFP was hydrophobic and floated on deionized water, whereas treated CFP was hydrophilic and sank immediately. **B** Agitating untreated CFP in water and pushing it under did not make untreated CFP sink, untreated CFP instantly resurfaced and floated.