

# 1 **Optimizing electrochemical degradation of** 2 **perfluorooctanoic acid in landfill leachate using** 3 **ceramic carbon foam electrodes by coupling CFD** 4 **simulation and reactor design**

5 Mattia Pierpaoli<sup>a</sup>, Pawel Jakóbczyk<sup>a</sup>, Małgorzata Szopińska<sup>b</sup>, Jacek Ryl<sup>c</sup>, Chiara Giosué<sup>d</sup>,  
6 Maciej Wróbel<sup>a</sup>, Gabriel Strugala<sup>e</sup>, Aneta Luczkiewicz<sup>b</sup>, Sylwia Fudala-Książek<sup>f</sup>, Robert  
7 Bogdanowicz<sup>a</sup>

8 <sup>a</sup> Faculty of Electronics, Telecommunications and Informatics, Department of Metrology and Optoelectronics,  
9 Gdańsk University of Technology, 11/12 Gabriela Narutowicza Street, 80-233 Gdańsk, Poland;  
10 [mattia.pierpaoli@pg.edu.pl](mailto:mattia.pierpaoli@pg.edu.pl) (MP, *Corresponding Author*, Tel +48 58 3471503 Fax +48 58 3471848)

11 [pawel.jakobczyk@pg.edu.pl](mailto:pawel.jakobczyk@pg.edu.pl) (PJ) [maciej.wrobel@pg.edu.pl](mailto:maciej.wrobel@pg.edu.pl) (MW) [robbogda@pg.edu.pl](mailto:robbogda@pg.edu.pl) (RB)

12 <sup>b</sup> Faculty of Civil and Environmental Engineering, Department of Environmental Engineering Technology Gdańsk  
13 University of Technology, 11/12 Gabriela Narutowicza Street, 80-233 Gdańsk, Poland; [malszopi@pg.edu.pl](mailto:malszopi@pg.edu.pl) (MS)  
14 [ansob@pg.edu.pl](mailto:ansob@pg.edu.pl) (AL)

15 <sup>c</sup> Faculty of Applied Physics and Mathematics, Division of Electrochemistry and Surface Physical Chemistry,  
16 Gdańsk University of Technology, 11/12 Gabriela Narutowicza Street, 80-233 Gdańsk, Poland; [jacryl@pg.edu.pl](mailto:jacryl@pg.edu.pl)  
17 (JR)

18 <sup>d</sup> Department of Materials, Environmental Sciences and Urban Planning (SIMAU), Università Politecnica delle  
19 Marche, Via Breccia Bianche 12, 60131 Ancona, Italy; [c.giosue@univpm.it](mailto:c.giosue@univpm.it) (CG)

20 <sup>e</sup> Department of Materials Science and Technology, Institute of Manufacturing and Materials Technology, Faculty  
21 of Mechanical Engineering and Ship Technology, Gdańsk University of Technology, 11/12 Gabriela Narutowicza  
22 Street, 80-233 Gdańsk, Poland; [gabriel.strugala@pg.edu.pl](mailto:gabriel.strugala@pg.edu.pl) (GS)

23 <sup>f</sup> Faculty of Civil and Environmental Engineering, Department of Sanitary Engineering, Gdańsk University of  
24 Technology, 11/12 Gabriela Narutowicza Street, 80-233 Gdańsk, Poland. [sksiazek@pg.edu.pl](mailto:sksiazek@pg.edu.pl) (SFK)

25

## 26 **Abstract**

27 Perfluorooctanoic acid (PFOA), a persistent and bioaccumulative pollutant classified as a  
28 'forever chemical', poses a global environmental and health risk due to its widespread use and  
29 resistance to degradation. The development of effective and efficient removal technologies is  
30 crucial to mitigate its long-term impacts. In this study, we present a novel approach to address  
31 the growing concern of emerging contaminants, particularly PFOA, in landfill leachate. We  
32 investigate the use of ceramic carbon foam electrodes (CCFE) as a cost-effective and efficient  
33 alternative to traditional electrode materials for the electrochemical degradation of PFOA.  
34 Micro-computed tomography was used to reconstruct the actual three-dimensional geometries  
35 of the samples in which porosities were calculated. We also coupled computational fluid  
36 dynamics simulations of the actual geometries and optimised the working conditions to

37 minimise pressure drop and improve treatment efficiency. Our design significantly reduces  
38 energy requirements by operating at lower potentials, a critical factor in sustainable waste  
39 management practices. The optimised CCFE system demonstrated superior performance in  
40 the degradation of PFOA in landfill leachate, offering a promising solution for the treatment of  
41 emerging contaminants. This study not only provides a viable method for mitigating the  
42 environmental impact of PFOA, but also sets a precedent for the development of low-energy,  
43 high-efficiency treatment technologies for various persistent pollutants. In addition, the  
44 proposed solution, as part of closed-loop water systems, will enhance water reuse and  
45 recycling, thereby preserving and regenerating natural water bodies.

## 46 **Keywords**

47 PFAS, electrochemical oxidation, advanced oxidation process, electrode stability

48

## 49 **1. Introduction**

50 The global increase in the volume and complexity of waste poses serious risks to ecosystems  
51 and human health (Clift et al., 2018; Falandysz et al., 2022). These growing waste streams are  
52 a combined source of emerging contaminants, including per- and polyfluoroalkyl substances  
53 (PFAS) (Reinhart et al., 2023). Due to the strong carbon-fluorine (C-F) bond, the persistent  
54 nature of PFAS allows them to persist in the environment, accumulate in soil and water, and  
55 thereby enter the food chain (Wee and Aris, 2023; Zafeiraki et al., 2019), raising significant  
56 health concerns. These concerns include the potential for both chronic and acute adverse  
57 human health effects, such as developmental problems, liver damage and an increased  
58 likelihood of cancer (Goodrich et al., 2022), as well as effects on wildlife (De Silva et al., 2021;  
59 Zafeiraki et al., 2019). Although many environmental contaminants are regulated and  
60 managed, effective control measures for PFAS and similar emerging contaminants remain  
61 inadequate. In particular, there is an urgent need for cost-effective technologies to remove  
62 persistent contaminants before they are released to the environment (Brunn et al., 2023;

63 Falandysz et al., 2022), especially from landfill leachates (Pierpaoli et al., 2021b). Efforts to  
64 remove PFAS from water include activated carbon adsorption and various physical and  
65 chemical treatments such as reverse osmosis, nanofiltration, ultrafiltration, oxidation and ion  
66 exchange, as well as biological treatments including biodegradation and constructed wetlands  
67 (EUN et al., 2022; Lu et al., 2023). Despite their effectiveness, these treatments are costly and  
68 result in PFAS-containing waste, which poses disposal challenges. Innovative technologies,  
69 such as electrochemical methods, offer promising alternatives for the treatment of PFAS-  
70 contaminated waters (Pierpaoli et al., 2021b; Xiao et al., 2023). These methods are valued for  
71 their versatility and cost-effectiveness, using electrons to directly or indirectly oxidise  
72 contaminants. A major focus is the use of three-dimensional electrodes, which come in various  
73 forms such as meshes, foams and porous solids, and are effective in increasing the rate of redox  
74 reactions. The movement of mass towards the electrode surfaces involves migration,  
75 convection and diffusion. The role of mixing in this context is twofold. Firstly, it helps to  
76 uniformise the concentration of reactants within the solution, ensuring a consistent and  
77 homogeneous mixture; secondly, it helps to reduce the thickness of the Prandtl layer  
78 immediately adjacent to the electrode surface.

79 The adsorption and electrochemical oxidation (EO) of perfluorooctanoic acid (PFOA) using  
80 unconventional materials have received considerable attention due to their low cost, high  
81 efficiency and environmental friendliness. While no adsorption of PFOA was observed on the  
82 negatively charged silica surface, PFOA and other PFASs were found to sorb to the alumina  
83 surface (positively charged), forming a hydrated layer, suggesting that the adsorption process  
84 appears to be driven by electrostatic interactions between the PFAS functional group (negative)  
85 and the mineral surface (Hellsing et al., 2016). Hydrotalcite is a layered double hydroxide  
86 (LDH) with the general formula  $Mg_6Al_2CO_3(OH)_{16-4H_2O}$  and has been shown to have  
87 extraordinary adsorption capacities for PFOA, of the same order as anionic exchange resins  
88 (above 100 mg/g) (Alonso-de-Linaje et al., 2021). Similarly, Wang and Shish showed how  
89 PFOA adsorption on crystalline alumina is influenced by the pH of the solution and how it  
90 decreases with increasing ionic strength of the cations, due to the compression of the electrical

91 double layer, as both Ca and Mg can form bridges with PFOA anions in solution (Wang and  
92 Shih, 2011). In fact, PFOA adsorption on inorganic oxide materials is mostly driven by  
93 electrostatic interactions (Chen et al., 2021). Al oxides and hydroxides, such as those derived  
94 from aluminium-based drinking water treatment residuals, have also been shown to strongly  
95 adsorb PFOA, with the capacity decreasing with increasing pH (Zhang et al., 2021). Open-cell  
96 foams have attracted considerable attention for various applications, such as catalytic  
97 substrates in combustion, reformers and converters to control pollutant emissions. They have  
98 also been of interest in the energy sector, particularly for heat transfer applications and as  
99 catalytic substrates in combustion devices, fuel reforming systems and emission control. The  
100 appeal of open-cell foams lies in their light weight, high surface to volume ratio and excellent  
101 fluid permeability, making them suitable for a wide range of engineering applications. The  
102 microstructural geometry of open-cell foams induces a tortuous flow path, resulting in high  
103 activity per unit volume, which is particularly advantageous for the aforementioned  
104 applications. Researchers have used CFD simulations to study flow regimes within ideal foam  
105 structures and the transition from laminar to fully turbulent regimes. Indeed, understanding  
106 the pressure drop and flow type as a function of flow rate within such a porous structure is  
107 crucial. The Reynolds number has been used to define the flow regime, with smaller pore sizes  
108 resulting in lower Reynolds numbers, leading to laminar flow.

109 The aim of our study was to explore the use of ceramic carbon foam electrodes (CCFE) as a  
110 cost-effective replacement for traditional electrode materials in the electrochemical  
111 degradation of PFOA found in landfill leachate. We focused on selecting a more affordable  
112 material and free from critical raw materials (CRMs) compared to traditionally available  
113 options such as boron-doped and metal oxide-coated titanium electrodes (Pierpaoli et al.,  
114 2020). Additionally, we optimized the flow and operational conditions using computational  
115 fluid dynamics (CFD) simulations, with a particular emphasis on minimizing energy  
116 consumption.

## 117 2. Materials and methods

### 118 2.1 Chemicals and Materials

119 Perfluorooctanoic acid (PFOA) standard, an analytical standard grade, was purchased from  
120 DR Ehrenstorfer (Germany). Eluent additive to HPLC method: ammonium acetate was  
121 suitable for mass spectrometry (LiChropur™, Merck Company), while methanol (MeOH), was  
122 hypergrade for LC-MS (LiChrosolv®, Merck Company). Phosphate buffer solution (PBS, 0.1  
123 M) was prepared as described in our previous work (Pierpaoli et al., 2021b). Purified water  
124 (ultrapure water, resistivity 18.2 MΩ cm at 25°C) was obtained via a Direct-Q® Water  
125 Purification System, and used to dilute 1:1 the landfill leachate and spike at different  
126 concentrations (0.001, 0.1 and 10 mg L<sup>-1</sup>) with PFOA. Ceramic carbon foam electrodes (CCFE)  
127 were supplied by Ferro-Term (Poland). Briefly, CCFE were prepared from a mixture of ceramic  
128 powder, hard coal and carbon binder, suspended in an aqueous silicic acid sol solution. The  
129 solution is then used to coat polymer foams, which, after drying, are fired at a temperature of  
130 600-1000°C in an oxygen-free atmosphere.

### 131 2.2 CCFE Chemical and morphological characterization

132 Scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) spectroscopy were  
133 performed using a Phenom XL microscope (Thermo Fisher Scientific) with a beam acceleration  
134 voltage of 10 kV, operating in high vacuum mode and with a secondary electron detector (SED).  
135 Raman spectroscopy was used to study the molecular composition of CCFE. The system  
136 consists of a TE-cooled to -60° CCD camera detector (Andor iDus-420-BR-DD, Ireland), a  
137 f/2.2 spectrograph with volume phase holographic (VPH) transmission grating (HT-  
138 spectrometer, EmVision llc., USA) with dedicated fibre-optic contact probe (EmVision llc.,  
139 USA) and an excitation laser module with central wavelength of 830 nm (Innovative Photonic  
140 Solutions, USA). Micro-computed tomography (μCT) was used for 3D reconstruction of the  
141 actual geometry of the samples. The equipment used was a GE phoenix vltomelxs with an X-  
142 ray power set at 30 W (100 kV, 300 μA) and an exposure time of 5000 ms for each radiogram.  
143 Additional information on μCT are reported in the supplementary materials. Two copper  
144 filters, each with a thickness of 0.5 mm, were used. ICP-OES (Inductively Coupled Plasma -

145 Optical Emission Spectrometer, Optima 8300 PerkinElmer) analyses were performed on the  
146 PFOA solutions, on the LL solution (after 30 minutes the CCFE electrode was in contact with  
147 the spiked LL and after full EO test). A constant voltage was then applied and the test was run  
148 for 3 hours. The samples collected were 10 mL of liquid for each analytical step. Prior to  
149 analysis, the samples were filtered at 45  $\mu\text{m}$  and 0.1 ml of concentrated  $\text{HNO}_3$  (ACS acid grade  
150 at 65%) was added to prevent precipitation of metals. Each CCFE was used once.

### 151 2.3 CCFE Electrochemical characterization

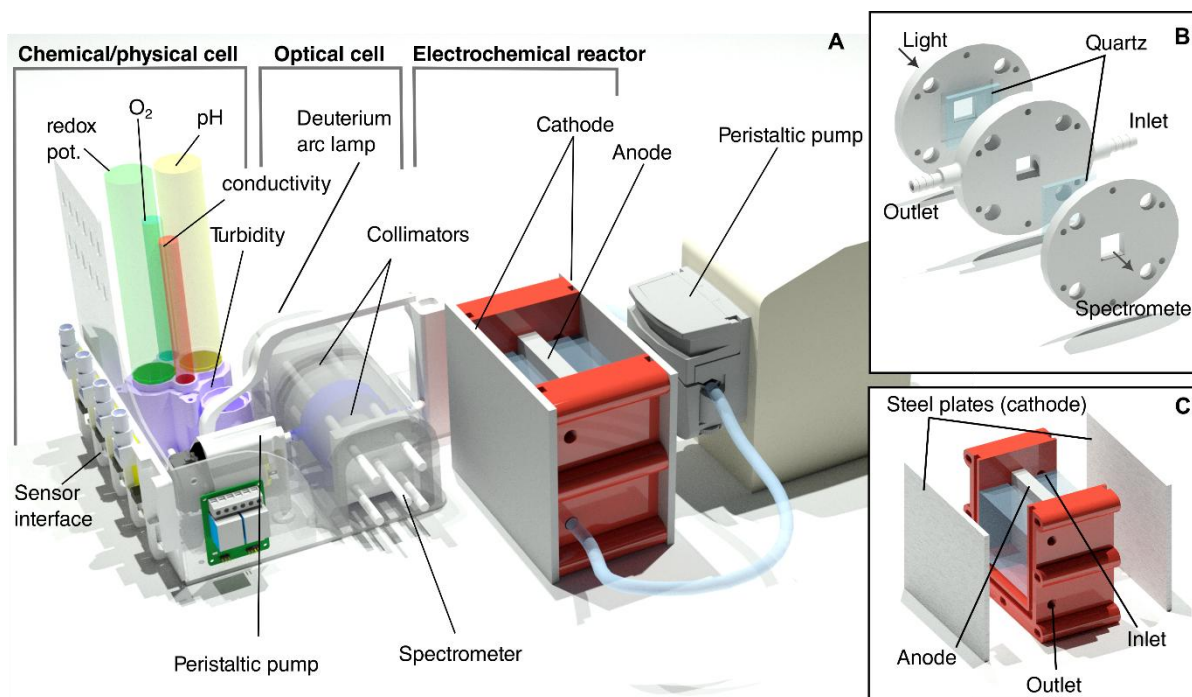
152 Electrochemical measurements, including Tafel plots, electrochemical impedance  
153 spectroscopy (EIS), and potential versus current, were performed before and after electrolysis  
154 in a three-electrode system in a beaker with a working electrode (CCFE) measuring 7.5 x 7.6 x  
155 2.5 cm, while the counter (cathode) and reference electrodes were a steel grid and Ad/AgCl  
156 wire, respectively. Tafel measurements were recorded from 0 to 4 V at a scan rate of 10  
157 mV/min. A current-potential curve for the CCFE in LL was obtained in the range of -1.5 V to 4  
158 V at a scan rate 20 mV/s. EIS was performed in the frequency range from 1 MHz to 100 mHz,  
159 at open circuit potential and with an amplitude of 20 mV. Electrochemical measurements were  
160 carried out with a potentiostat-galvanostat (VMP-300, Bio-Logic, France) controlled by EC-  
161 Lab software. The electrolyte used for electrochemical measurements was 0.1 M PBS.

162

### 163 2.4 Reactor and monitoring system design

164 The electrochemical reactor and monitoring cell were designed and realized by additive  
165 manufacturing in PETG. Monitoring cell geometry was optimized by CFD simulation, as well  
166 as the residence time was estimated to minimize the time between two subsequent samples.  
167 The monitoring cell includes 4 low-cost Electrical Conductivity, dissolved  $\text{O}_2$  (DO), redox  
168 potential, pH sensors (Table S1), which have been calibrated against a laboratory grade digital  
169 multimeter (HQ40D, Hach), equipped with HQ2200 (pH); IntelliCAL LDO101 (DO); PT1000  
170 (conductivity) and IntelliCAL MTC101 (redox) probes manufactured by Hach (Germany). The  
171 optical cell (Fig. 1B) includes a small cell (10 mm x 10 mm and 6 mm of optical path length)  
172 comprised of two quartz glasses. From one of the two sides, the water sample was illuminated

173 with a halogen source through an optical fibre and a collimator, and the resulting spectra were  
 174 passed through collimator and an optical fibre and acquired using a portable UV-VIS  
 175 spectrometer (Ocean Optics FX). For the monitoring and optical cells, two peristaltic pumps  
 176 regulated the flow. All optical and chemical measurements were conducted under static  
 177 conditions, meaning there was no flow during these measurements. Leachate was directed to  
 178 the measurement cells at five-minute intervals for sensor analysis. Additionally, samples taken  
 179 at specific time intervals - 0, 5, 15, 30, 60, 90, 120, and 150 minutes - were specifically  
 180 designated for analysis using ultra-high performance liquid chromatography tandem mass  
 181 spectrometry with electrospray ionisation (UHPLC-ESI-MS/MS). The electrochemical reactor  
 182 was designed as a parallelepiped chamber to facilitate flow through the CCFE electrode, with  
 183 the inlet and outlet positioned at diametrically opposed openings in the reactor (Fig. 1C). For  
 184 each experiment, the system was allowed to stabilise for 30 min before a constant voltage was  
 185 applied. This stabilisation period ensured that the adsorption on the electrode reached a steady  
 186 state. A constant potential was then maintained for 180 minutes in each experiment, with the  
 187 current measured continuously throughout the process.



188

189 *Fig. 1. – (A) Setup for the EO tests; (B) Optical cell; (C) EO reactor*

190 The energy consumption per unit mass of PFOA removed ( $EC_{PFOA}$ ) for the electrochemical  
 191 mineralization process at a giving reaction time  $t$  (h) was calculated as [29]:

$$192 \quad EC_{PFOA} = \frac{E_{cell}It}{\Delta[PFOA]_tV} \quad \text{Eq. 1}$$

193 where  $E_{cell}$  is the cell potential (V),  $I$  the applied electric current (A),  $\Delta[PFOA]_t$  ( $\text{mg L}^{-1}$ ) the  
 194 variation of [PFOA] after a certain time  $t$  (h), and  $V$  the solution volume (L).

## 195 2.5 Adsorption experiments

196 Adsorption isotherm experiments were carried out in 120 mL HDPE vessels containing 100  
 197 mL solution. The initial concentrations of PFOS varied from  $0.02 \text{ mg L}^{-1}$  to  $0.5 \text{ mg L}^{-1}$  and from  
 198  $10 \text{ mg L}^{-1}$  to  $125 \text{ mg L}^{-1}$  to achieve two adsorption isotherms at low and high concentrations. The  
 199 vessels were shaken for 24 h. The amount of PFOS adsorbed by the CCFE and the single-point  
 200 adsorption coefficients  $K_d$  ( $\text{L g}^{-1}$ ) were calculated from

$$201 \quad q_t = \frac{C_0 - C_t}{C_s} \quad \text{Eq. 2}$$

$$202 \quad K_d = \frac{q_e}{C_e} \quad \text{Eq. 3}$$

203 where  $q_t$  and  $q_e$  ( $\mu\text{mol g}^{-1}$ ) are the concentrations of solutes adsorbed on the adsorbents at time  
 204  $t$  (min) and at equilibrium, respectively.  $C_0$  ( $\mu\text{mol L}^{-1}$ ) is the initial solute concentration.  $C_t$  and  
 205  $C_e$  ( $\mu\text{mol L}^{-1}$ ) are the solute concentrations in the aqueous phase at time  $t$  and at equilibrium,  
 206 respectively.  $C_s$  ( $\text{g L}^{-1}$ ) is the mass of the CCFE.

207 The solid–liquid sorption distribution coefficient,  $K_d$  ( $\text{mL g}^{-1}$ ), was calculated as the ratio  
 208 between the PFOA concentration sorbed in CCFE,  $C_s$  ( $\text{mg g}^{-1}$ ), and its concentration in the  
 209 aqueous phase at equilibrium,  $C_{eq}$  ( $\text{mg L}^{-1}$ ):

$$210 \quad K_d = \frac{C_s}{C_{eq}} \quad \text{Eq. 4}$$

211  $C_{eq}$  values were directly determined by UPLC–MS/MS and the corresponding concentrations  
 212 of PFASs sorbed by CCFE, whereas  $C_s$  was calculated by the following equation:

$$213 \quad C_s = \frac{(C_{in} - C_{eq})V}{m_{CFE}} \quad \text{Eq. 5}$$



214 where  $C_{in}$  ( $\text{mg L}^{-1}$ ) corresponds to the initial concentration of PFOA added to the CCFE,  $V$  (mL)  
215 is the volume of solution and  $m_{CCFE}$  (g) refers to the dry mass of the CCFE.

216 The Freundlich model is quantitatively described by the following equation:

$$217 \quad C_s = K_f C_{eq}^n \quad \text{Eq. 6}$$

218 Where  $K_f$  is the Freundlich constant, related to the adsorption capacity of the adsorbent and  
219 the intensity of adsorption and  $n$  is the Freundlich exponent, a dimensionless constant that  
220 represents the heterogeneity of the adsorption sites on the solid surface. The  $K_f$  and  $n$   
221 parameters of the Freundlich equation were calculated by non-linear least squares fitting of  
222 the sorption isotherms.

## 223 2.6 UHPLC – MS/MS and chemical analysis

224 An UHPLC-ESI-MS/MS Nexera XR coupled with LC/MS-8050 (Shimadzu Company), was  
225 used to determine PFOA concentration. Process control samples were filtrated via  
226 polypropylene (PP) 0.45  $\mu\text{m}$  pore size syringe filters (Whatman®) and directed to UHPLC-  
227 ESI-MS/MS analysis. Modified and adjusted to the study purposes EPA 537.1 method was  
228 applied. Shim-pack SP-C18, 2.1 x 150 mm, and 2.7  $\mu\text{m}$  was used as an analytical column, with  
229 a constant flow applied at 0.3  $\text{ml min}^{-1}$ . The sample injection volume was 1 or 5  $\mu\text{L}$  (depending  
230 on the spiking level). The calibration curve was plotted using 5-level calibration (linearity in  
231 the ranges of 1–100  $\mu\text{g L}^{-1}$  and 1–10  $\text{mg L}^{-1}$ ). The method detection limit (MDL) and method  
232 quantification limit (MQL) were 1 and 3  $\mu\text{g L}^{-1}$  for the landfill leachate matrix. For this method,  
233 no analyte pre-concentration was applied. Additional details are reported in the  
234 supplementary materials. The composition of the landfill leachate was analysed using the  
235 methods described in detail in our previous work (Fudala-Ksiazek et al., 2018; Szopińska et al.,  
236 2021; Wilk et al., 2022).

## 237 2.7 CFD simulation

238 A comprehensive review on modelling turbulent flows in porous media, among which metal  
239 foams, has been done by Wood, He and Apte (Wood et al., 2020). Briefly, in porous media,  
240 four distinct flow regimes can be identified: (a) Darcy, (b) Forchheimer, (c) unsteady laminar



241 (transitional), and (d) fully-turbulent regime (Dybbs and Edwards, 1984). The transition  
 242 between these regimes is described by the pore-based Reynolds number, given by the formula:

$$243 \quad Re_p = \frac{UL_p}{\nu} \quad \text{Eq. 7}$$

244 In which  $L_p$  is assumed equal to the diameter of the average pore size,  $U$  is the intrinsic average  
 245 velocity and  $\nu$  the kinematic viscosity.

246 Firstly, by considering a 20 x 20 x 60 mm representative elementary volume (REV), steady  
 247 Reynolds-averaged Navier–Stokes (RANS) simulations were performed at various Re number.  
 248 The REV was obtained by  $\mu$ CT of the real sample and subsequent the mesh was reconstructed  
 249 by voxel segmentation. Meshing ( $4.5 \times 10^6$  elements) and CFD simulations were performed with  
 250 COMSOL Multiphysics 6.1.

251 The  $v^2$ -f model (Durbin, 1991), a higher order turbulence model, suggested to be particularly  
 252 suitable for turbulent flow in coarse porous media (Kazerooni and Hannani, 2007), has been  
 253 chosen. Then CFD simulation at different flow rates have been performed, in order to obtain  
 254 the averaged values for permeability to describe the macroscopic model. Permeability,  $\kappa$ , is  
 255 given by the Eq. 8

$$256 \quad \kappa = u_{out} \mu \frac{L}{\Delta p} \quad \text{Eq. 8}$$

257 Where  $\mu$  is the dynamic viscosity,  $u_{out}$  the outlet fluid velocity in the flow direction,  $L$  the side  
 258 length and  $\Delta p$  the pressure drop across the REV.  $\kappa$  has been calculated for low velocities  
 259 (equivalent to  $Re_p < 12$ ), for which we found a linear relation was still valid.

260 Secondly, the reactor reported in Fig. 1c has been simulated, using a porous media  
 261 characterized by porosity  $\varepsilon_p$  and permeability  $\kappa$  as the CCFE with the additional Forchheimer  
 262 coefficient equal to:

$$263 \quad \beta = \frac{\rho \varepsilon_p C_f}{\sqrt{\kappa}} \quad \text{Eq. 9}$$

264 Having  $\rho$  the fluid's density and  $C_f$  the dimensionless Friction coefficient (Amiri and Vafai,  
 265 1998), equal to 0.23 (Amiri and Vafai, 1998).

266



### 267 **3. Results and discussion**

#### 268 **3.1. Landfill leachate characterization**

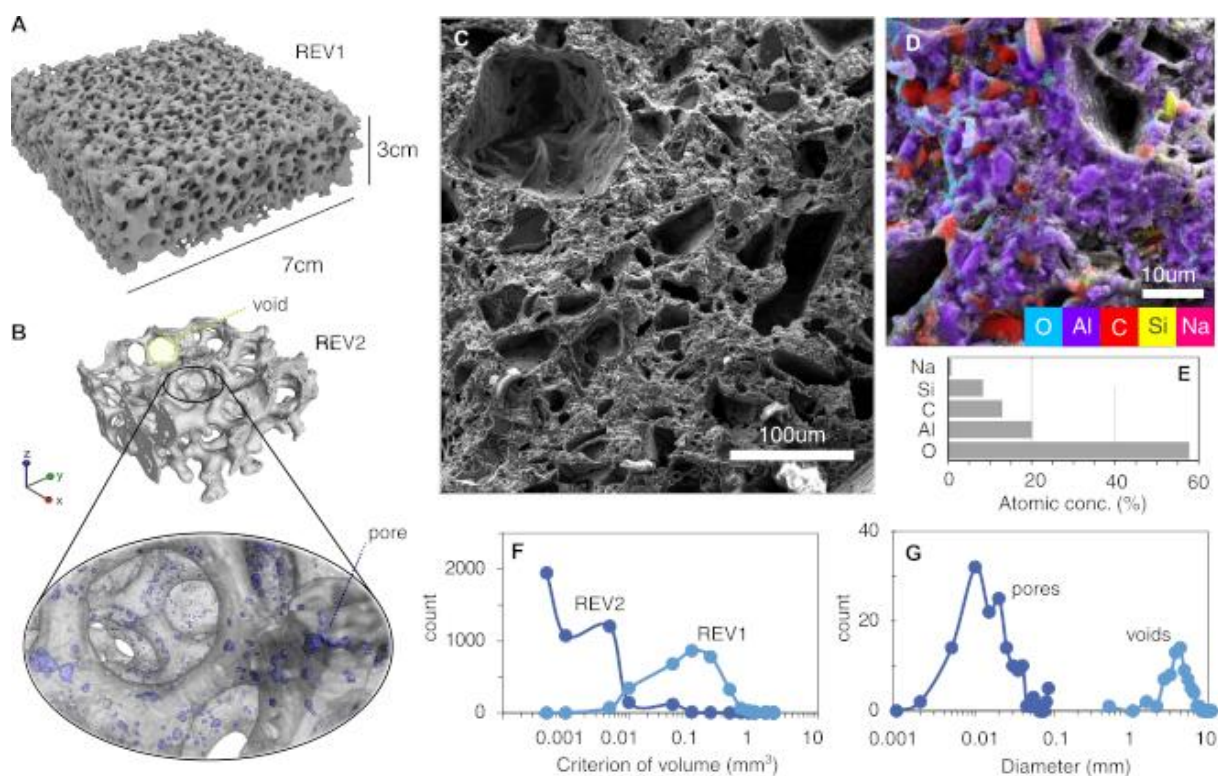
269 The landfill leachate (LL) was taken from the municipal solid-waste plant (MSWP) 'Eko Dolina  
270 Lezyce' located in the Pomerania region (northern Poland) from the cell operated during a  
271 period where there was no waste segregation (from 2003 to 2011). This may cause various  
272 micropollutant presence in the samples (including contaminants from the perfluorinated  
273 organic compounds). The samples of raw LL were collected to polyethylene bottles in August  
274 2022 and transported to the laboratory (at  $4 \pm 1^\circ\text{C}$ ), where their physicochemical properties  
275 were immediately evaluated. The basic composition of the landfill leachate is summarised in  
276 Table S2.

#### 277 **3.2. Setup characterization and parameter optimization**

278 CCFE is characterized by a three-dimensional connected mesh structure and high porosity,  
279 small flow resistance and it is electrically conductive. The fabrication of CCFE is conducted via  
280 a multi-stage procedure. The process starts with the application of a ceramic suspension,  
281 comprising alumina, sintering coal, and a blend of additives, onto an elastic polymer foam.  
282 This slurry is employed to impregnate the polymeric foam, which acts as a foundational  
283 skeleton for the ensuing structure. Following the application of the slurry, the foam filters are  
284 subjected to a drying phase. Then, filters are placed in a kiln and exposed to elevated  
285 temperatures. During the firing phase, the organic skeleton within the filters is incinerated,  
286 resulting in an interconnected, porous ceramic matrix. This structure is then sintered to  
287 enhance its mechanical strength and durability. Concurrently, caking coal, is added. The  
288 heating process induces the caking coal to initially attain a plastic state, facilitating its  
289 adherence to and encapsulation of the ceramic component grains. As the temperature  
290 increases, the caking coal undergoes sintering, forming a sturdy ceramic-carbon composite  
291 material (Aslanowicz et al., 2019; Karwiński et al., 2013).

292 The resulting solid matrix is a heterogenous composite rich of Al, C, Si oxides (Fig. 2D,E). Fig.  
293 2A presents the 3D reconstruction of the microtomography of the entire CCFE (REV1)  
294 employed in the experiments. For more detailed observations of inner pores, a smaller sample

295 (REV2) was reconstructed at a higher resolution, as illustrated in Fig. 2B. In addition, the  
 296 diameters of inner pores and voids were analysed based on microphotographs taken by SEM  
 297 (Fig. 2C). From SEM/EDX analysis (Fig. 2D) it is possible to see that the composition of the  
 298 raw CCFE accounts mostly of Al, C, Si and Na (Fig. 2E). The total porosity is a complex outcome  
 299 influenced by the sacrificial polymeric structure, which is skeletal and characterized by a high  
 300 void content, along with the presence of inner pores. The volume of inner pores has been  
 301 calculated for both REV samples, and the distribution is depicted in Fig. 2F. The inner pore  
 302 diameters, shown in Fig. 2G, were in the range of tens of micrometers, and the distribution of  
 303 void diameters, as shown in the same figure, had a median diameter of approximately 3.8 mm.  
 304 Full porosity calculations for both samples based on 3D reconstructions are shown in Table 1.  
 305 The total porosity of the REV1 sample was 72.4%, while that of the REV2 sample was 79.6%.  
 306 The differences in porosity between the samples are attributed to the different volumes used  
 307 for calculations and the different resolutions (voxel size) obtained. In addition, the surface area  
 308 of the samples that were counted is given in Table S3.



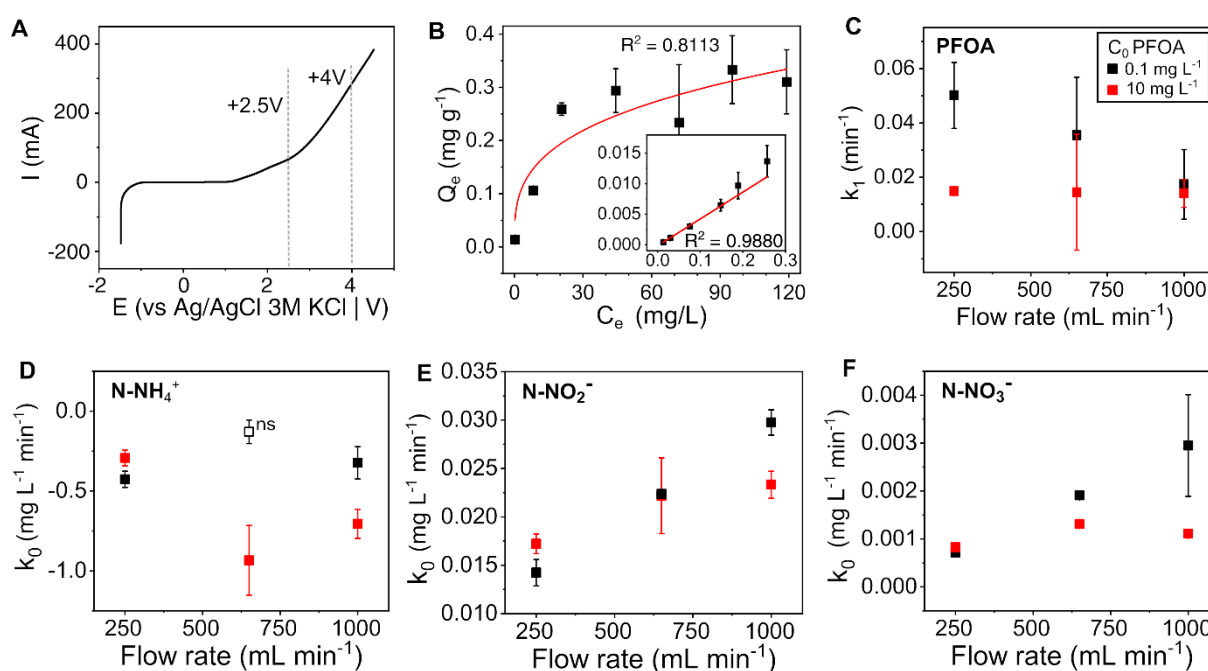
309  
 310 *Fig. 2. – A) 3D visualization of a reconstructed larger sample; B) 3D visualization of a reconstructed*  
 311 *smaller sample with marked void and a magnified semi-transparent image showing pores; C) Cross-*

312 section of sample observed by SEM; D) Map of elements on the cross-section of the sample observed  
 313 by SEM and EDX; E) Elemental analysis of the sample; F) Volume distribution of pores and (G)  
 314 diameter distribution of pores and voids of the REV's.

315

### 316 3.3. PFOA oxidation in landfill leachate

317 A current-potential curve for the CCFE in LL was obtained by scanning from -1.5 V to 4 V (vs  
 318 Ag/AgCl 3M KCl). From Fig. 3A it is possible to observe two linear trends from which two  
 319 potentials, 2.5 V and 4 V, have been selected to perform the EO experiment under  
 320 potentiostatic conditions.



321 Fig. 3. - A) Potential vs current for CCFE in LL; B) PFOA adsorption isotherms at high and low (inset)  
 322 concentrations for the CCFE electrode; C) PFOA degradation rate as function of the flow rate. Zero-  
 323 order rate constants for; D)  $N-NH_4^+$ ; E)  $N-NO_2^-$  and F)  $N-NO_3^-$  as function of the flow rate. "ns": not  
 324 significant for  $p > 0.05$ .  
 325

326

327 The adsorption of PFAS on various carbonaceous and inorganic materials has been extensively  
 328 described and reviewed elsewhere (Du et al., 2014; Valencia et al., 2023).  $C_s$  vs.  $C_{eq}$  plots were  
 329 used to construct sorption and then fitted to both Freundlich and linear models, which are  
 330 commonly used for hydrophobic compounds (Hinz, 2001) and have been previously applied to  
 331 describe PFAS sorption in sediments (Du et al., 2014; Helsing et al., 2016; Higgins and Luthy,

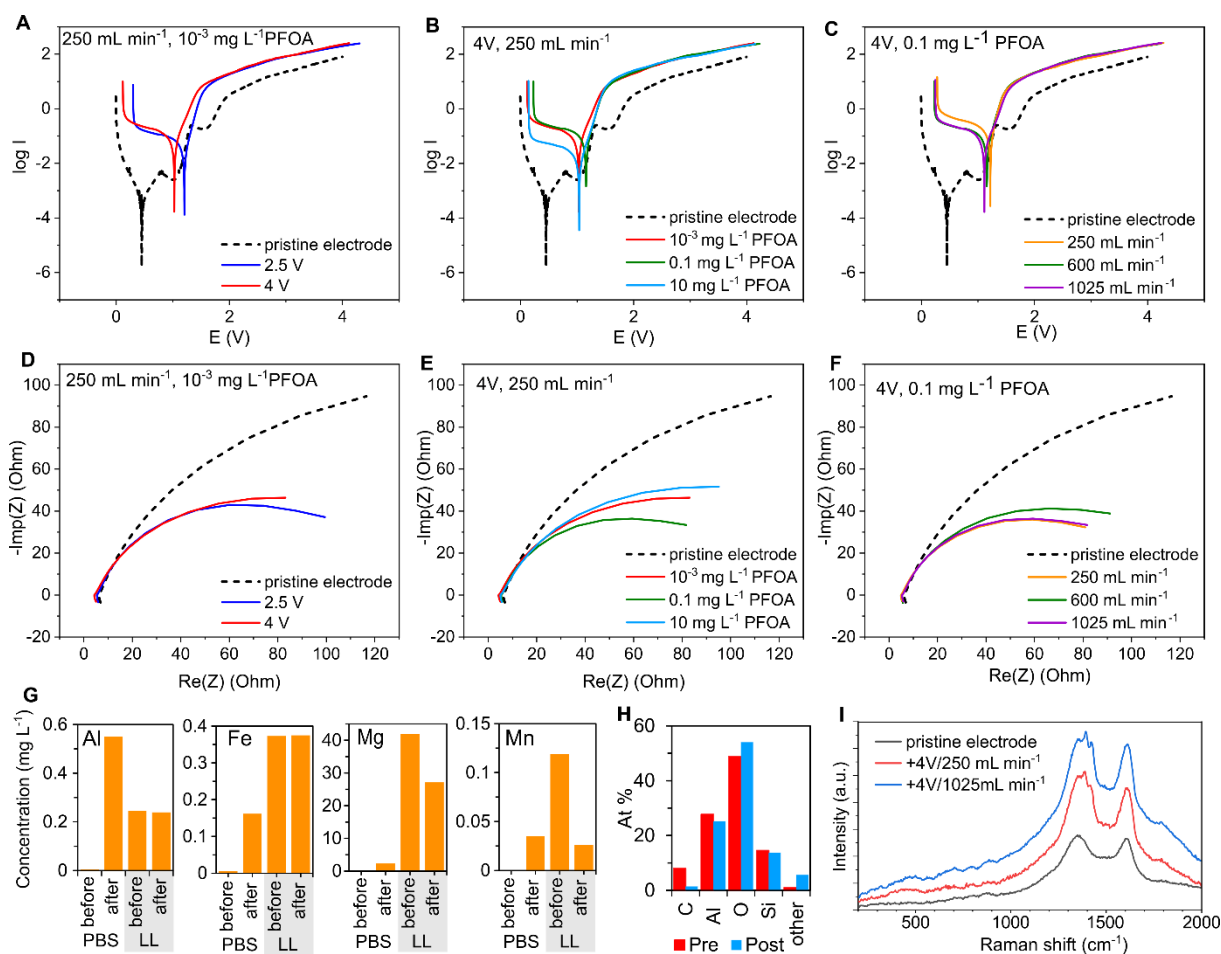
2006). For low PFOA concentrations (0.02-0.5 mg L<sup>-1</sup>), a linear relationship was observed with a  $K_d$  of 0.05 mL/mg, while for higher PFOA concentrations (10-125 mg L<sup>-1</sup>), the Freundlich isotherm fit the experimental results with a  $K_f$  of 0.078 mg<sup>(1-n)</sup>mL<sup>n</sup> g<sup>-1</sup> (Fig. 3B). A comparison with similar adsorbents by composition is given in Table S4. The first order rate constants of PFOA as a function of both initial concentration and flow rate are shown in Fig. 3C. It can be observed that at lower concentrations, the slower the flow through the CCFE, the higher the rate constant. While this may seem counterintuitive, since a higher flow rate implies a higher mixing rate, which has been reported in the literature to enhance PFOA oxidation (Asadi Zeidabadi et al., 2023), it also implies a lower residence time in the reactor and a lower contact time with the CCFE surfaces. Since PFOA oxidation is mostly driven by direct electron transfer, rather than the action of the  $\cdot$ OH radical (Pierpaoli et al., 2021b), where the limiting step is provided by the electrode active surface, the higher velocity through the CCFE results in a lower rate. Furthermore, we observed that no degradation occurs at the lower potential of +2.5 V (Fig. S1). A zero-order reaction was observed for ammonium removal, as it may be mainly driven by the indirect pathway through reaction with chlorine (Li and Liu, 2009), which is abundant in LL. Fig. 3D shows the variation of the ammonia rate constant as a function of the flow rate. While the  $\text{NH}_4^+$  concentration decreases as the EO process progresses, the  $\text{N-NO}_2^-$  and  $\text{N-NO}_3^-$  concentrations increase, also clearly at zero order, as ammonia is partially converted to nitrate ions (Anglada et al., 2011; Pierpaoli et al., 2021a). While a correlation between flow rate and  $\text{N-NH}_4^+$  removal rate is not visible, it is for the  $\text{N-NO}_2^-$  and  $\text{N-NO}_3^-$  rates, as they increase with increasing flow but they are less affected from the presence of PFOA, due to competitive direct oxidation on the surface electrode. The COD is slightly affected by the EO, since its degradation is accompanied by the release of carbon from the electrode (Fig. S2).

The optical transmittance of the LL was measured using the designed optical cell, with a halogen lamp as the source, in the ranges 350-550 nm and 670-950 nm. The 550-670 nm interval was discarded due to the presence of the lamp emission peak which saturated the spectroscope. When a potential of +4 V was applied, the transmittance in the range 400-550 nm decreased, which may be due to carbon loss from the electrode to the LL. Additionally, a

360 slight increase in transmittance was observed in the range 670-780 nm when a potential of  
361 +2.5 V was applied. An increase was also observed in the blank sample without the application  
362 of a potential. This increase is likely due to the sedimentation and/or adsorption of particles  
363 and compounds (Fig. S3). Fig. S4 shows the correlation between the parameters estimated by  
364 the chemical/physical monitoring cell. A general decrease in LL pH is observed, along with a  
365 slight increase in electrical conductivity. The oxidation reduction potential decreases abruptly  
366 from positive ( $84\pm 18$ )mV to negative ( $-66\pm 12$ )mV just 15 minutes after the application of the  
367 potential difference, and slowly increases with time to ( $-43\pm 24$ )mV at 150 minutes. Oxygen is  
368 rapidly depleted as it is consumed in the cathode, reaching its minimum after one hour of  
369 treatment. However, a higher flow rate supports the LL reoxygenation (see Fig. S5). The  
370 specific energy consumption for PFOA removal, as described in Eq. 1, decreases with  
371 increasing PFOA concentration, in a log-log correlation (Fig. S6).

### 372 3.4. Uniform corrosion rate and local corrosion susceptibility evaluation

373 To evaluate the susceptibility of uniform corrosion rate, the electrodes underwent cyclic  
374 polarization and electrochemical impedance spectroscopy measurements before and after an  
375 electrolysis event. Three external environmental factors were identified as important for  
376 material degradation rate and electrolytic efficiency: electrolyser polarization conditions,  
377 hydrodynamic fluid flow through the electrochemical cell, and the concentration of  
378 contaminant species in the electrolyte. It is important to note that even a single electrolysis  
379 cycle can lead to a deterioration in the corrosion resistance of the CCFE electrodes being  
380 studied. This can be observed through changes in the electrode's kinetics. Tafel extrapolation  
381 can be used to estimate the corrosion current ( $i_0$ ), which is directly proportional to the  
382 corrosion rate. Notably, the electrolysis voltage has a visible effect on the cathodic slope,  
383 indicating that both studied polarization conditions: +2.5 and +4.0 V lead to  $i_0$  increase by two  
384 orders of magnitude, higher at higher polarization voltages (Fig. 4.3A,D). The charge-transfer  
385 mechanism shifts from anodically controlled to cathodically controlled after electrolysis.



386  
 387 Fig. 4. – (A-C) cyclic polarization curves and (D-F) EIS spectra recorded for CCFE electrodes. (G) Al,  
 388 Fe, Mg, Mn concentrations, determined by ICP-OES, before and after both pre-conditioning in PBS  
 389 and in LL. (H) difference in elemental composition, determined by EDX and (I) Raman spectra of the  
 390 CCFE before and after EO tests.

391  
 392 It is known that some of the electro-oxidized species are adsorbing at the electrode surface,  
 393 acting as a passive film and effectively blocking the electrode towards consecutive oxidation  
 394 processes. This characteristic process may lead to a secondary decrease in electrode  
 395 susceptibility to corrosion, which should be explained by the passivation mechanism. As an  
 396 example, an effect of Perfluorooctanoic acid (PFOA) was used in different concentrations  
 397 ranging from 0.001 to 10 mg/L (Fig. 4B,E). Here, at the highest concentrations, the corrosion  
 398 current  $i_0$  was the lowest, which, however, has an expected negative influence on the electro-  
 399 oxidation efficiency. Notably, there was no shift in the corrosion potential observed, as would  
 400 be in the case of the classic passive layer. The CCFE corrosion susceptibility was studied at



401 various hydrodynamic flows, ranging from 250 to 1025 mL/min (Fig. 4.C,F). It is seen that  
402 regardless of the flow rate the mechanism of the corrosion process is quite similar, and the  
403 corrosion rate is not driven by the diffusion limitation. In other words, despite the flow rate  
404 studied the electrochemical process under investigation is activation-controlled, and the  
405 electron transfer is slower compared to the diffusion. This observation is connected to the foam  
406 shape of the studied electrodes. It should be noted that the lowest  $i_0$  was identified for the  
407 sample studied at the lowest flow rates.

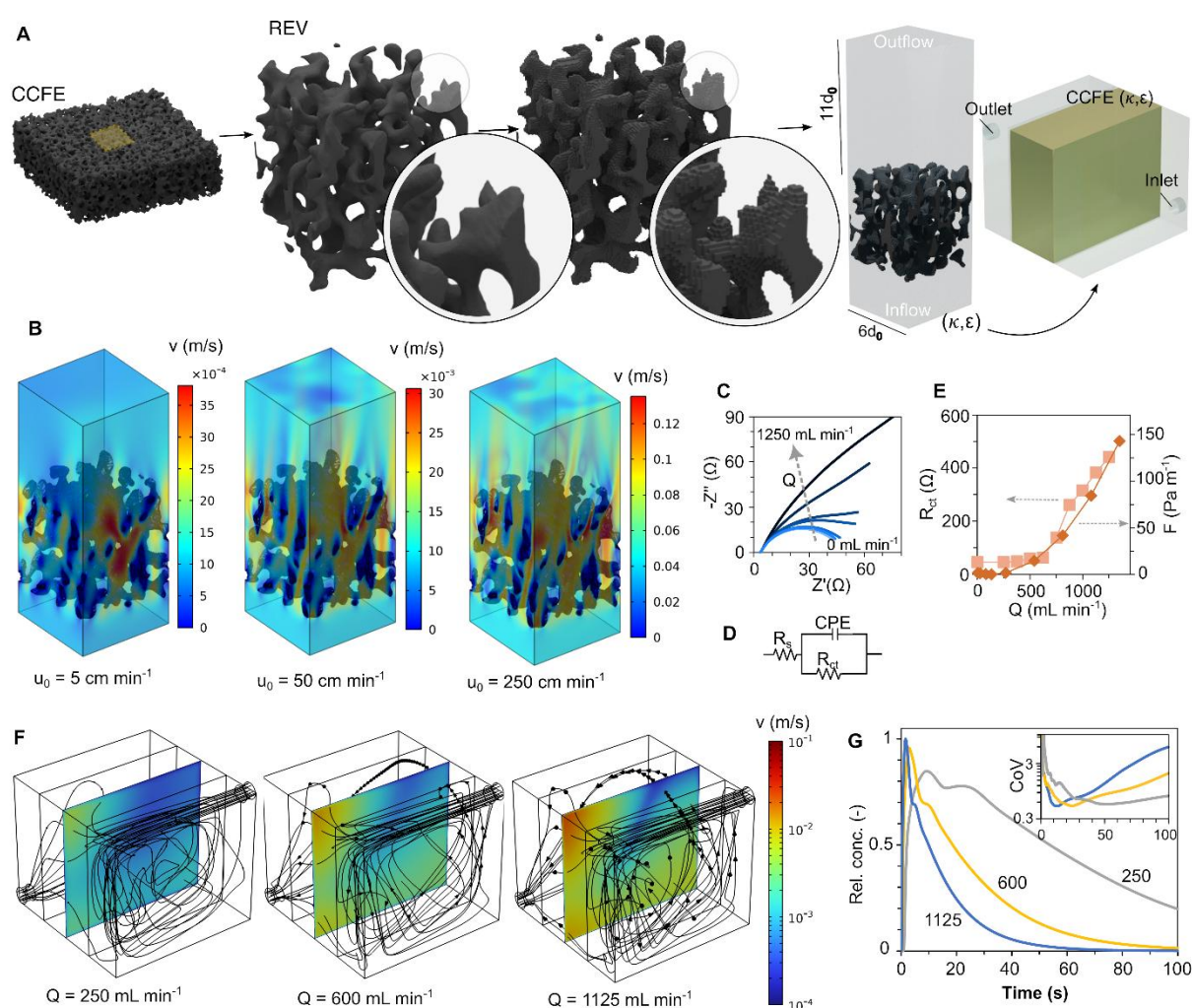
408 Part of the Al and Fe content of the CCFE is leached into the PBS prior the electrochemical  
409 oxidation test, during the Tafel characterization (Fig. 4G). Moreover, a decrease of Mg and Mn  
410 from LL are observed (Chen et al., 2022). The reason may be due the formation of magnesium  
411 hydroxide,  $Mg(OH)_2$ , especially in alkaline conditions, which can precipitate, since it becomes  
412 less soluble as the pH increases (Pilarska et al., 2017; Weijma et al., 2022); however, since the  
413 pH increases during the process, it may be possible that Mg may be adsorbed/electrodeposited  
414 on the electrode or in general, metals can have a rapid aggregation in aqueous solution due to  
415 large Van der Waals forces, and decreasing the adsorption potential (Qasem et al., 2021). In  
416 fact, according to the reduction at the cathode, the heavy metals solid gathers at the cathode  
417 and form a layer on its surface (Tran et al., 2017). Indeed, a reduction was detected on Cr and  
418 slightly on Ni (Fig. S7). Additionally, the EDX analysis reveals a loss of carbon content and an  
419 enrichment of other elements (Fig. 4H). The Raman spectra collected before and after the EO  
420 process show distinct peaks on top of the D peak, broadening of the D peak, and an increase in  
421 background fluorescence, possibly due to the loss of carbon from the Al-rich structure (Fig. 4I).  
422 The sharper peaks that arise are at approximately  $1395\text{ cm}^{-1}$  and  $1420\text{ cm}^{-1}$ , which most likely  
423 correspond to C–O stretching and  $CH_2$  scissor bending, respectively. Additionally, there is a  
424 weak, broad band near the G peak at about  $1760\text{--}90\text{ cm}^{-1}$ , which is likely attributable to C=O  
425 stretching, suggesting oxidation.

426 In summary, the most notable factor affecting the corrosion rate is the electrode modification  
427 under electrolysis, which leads to surface area development and modification of the redox

428 mechanism. The variation in polarization voltages and flow rates is noticeable but does not  
 429 exceed one order of magnitude.

### 430 3.5. CFD Simulation

431 The initial step in CFD simulation involved reconstructing the imported REV of the CCFE  
 432 to eliminate the internal porosity and simplify the geometry through castellation for import  
 433 into the software. After that, high-order turbulence model ( $v^2f$ ) has been employed to solve the  
 434 model. This helps obtain permeability and porosity values, which are then used in an  
 435 approximate model of the full reactor (Fig. 5A).



437 *Fig. 5. – (A) schematic representation of the procedure towards the EO reactor CFD simulation. (B)*  
 438 *Velocity magnitude across the REV for three selected inlet velocities. (C) EIS results at different flow*  
 439 *rates and (D) EEC used to fit the experimental data. (E)  $R_{ct}$  and  $F$  as function of the flow rate. (F)*  
 440 *Velocity magnitudes in the CCFE cross-section at half depth resulting from the simplified CFD*



441 *simulation of the EO reactor. (G) Relative concentrations within the CCFE volume as function of the*  
442 *time for an instantaneous release.*

443 Fig. 5B shows the change in the flow field through the CCFE porosity with increasing velocity.  
444 The CFD simulation investigated velocities ranging from 1 to 250 cm min<sup>-1</sup>. Meanwhile, EIS  
445 was performed on the CCFE electrode using the setup depicted in Fig. 1. The resulting  
446 impedance spectra at various flow rates (ranging from 250 to 1250 mL min<sup>-1</sup>) are shown in Fig.  
447 5C and fitted with the equivalent electrical circuit (EEC) presented in Fig. 5D. It is noteworthy  
448 that  $R_{ct}$  increases as the flow rate increases, specifically for  $Q > 625$  mL min<sup>-1</sup>, which is the  
449 threshold at which the Forchheimer contribution (Eq. 9) becomes relevant (Fig. 5D). The  
450 increased pressure drop and flow resistance associated with the Forchheimer term (F) affect  
451 the transport of species towards the electrode surface, which in turn affects charge transfer  
452 kinetics and subsequently the charge transfer resistance of the electrode. The simulation  
453 yielded values for porosity and permeability of 0.83 (similar to the total porosity ratio obtained  
454 by  $\mu$ CT) and  $6.4 \times 10^{-7}$  m<sup>2</sup>, respectively. These values were used to create a simplified model of  
455 the reactor. The flow is described by the Navier-Stokes equation in the free region and a  
456 Forchheimer-corrected version of the Brinkman equations in the CCFE region. The simulation  
457 results at the flow rates used in the EO tests are shown in Fig. 5F. An increasing heterogeneity  
458 of velocity intensity can be observed at the centre of the CCFE electrode as the flow rate  
459 increases. By simulating an instantaneous release of a given species at the inlet, it is possible  
460 to observe concentration peaks at the outlet at 4, 8, and 23 seconds for the three flow rates  
461 (Fig. 5G). The latter flow rate has the lowest coefficient of variation (calculated as Fig. S8) and  
462 is more constant over time (see inset), suggesting better mixing and concentration uniformity  
463 within the CCFE electrode.

#### 464 **4. Conclusions**

465 This study explores the use of CCFE for the electrochemical oxidation of PFOA in landfill  
466 leachate. The process parameters, such as applied potential and flow rate, were optimized, and  
467 an automated platform was developed to monitor the process, supported by CFD simulations.  
468 The PFOA adsorption onto CCFE is comparable to that on Al-rich sediments, and it provides a



469 valuable support for its electrochemical oxidation. Lower flow-through rates improve PFOA  
470 oxidation, while the opposite is true for the oxidation of N-NH<sub>4</sub> to N-NO<sub>3</sub> by N-NO<sub>2</sub>. PFOA  
471 follows a first-order kinetic due to direct electron transfer being the leading mechanism. In  
472 contrast, N-NH<sub>4</sub> oxidation is driven by the indirect reaction with the electrogenerated chlorine,  
473 resulting in a zero-order kinetic fashion due to the abundance of chlorides in LL. The results  
474 of the CFD simulation indicate that at lower flow rates, the concentration at the CCFE electrode  
475 cross-section is more uniform under Darcy flow conditions. Electrode stability was assessed  
476 using electrochemical methods, and the leaching of carbon and aluminium was confirmed by  
477 EDX and ICP-OES. Additionally, the removal of some metals in the solution, such as Mg and  
478 Mn, from LL and their possible deposition onto the electrode surface were detected by means  
479 of Raman investigations. By coupling spectroscopic methods with electrochemical ones, it is  
480 possible to observe that the transmittance through the LL increases at potentials up to 2.5 V  
481 without the oxidation of PFOA. However, at 4 V, PFOA is oxidised, resulting in a decrease in  
482 transmittance in the 480-550 nm interval. Additionally, CFD simulations can be used to  
483 optimize reactor geometry and operational parameters to develop a viable technology for PFAS  
484 control.

485

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