Supporting information

Phosphate recovery from human waste via the formation of hydroxyapatite during electrochemical wastewater treatment

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Number of pages: 13 (including this cover page)

Number of figures: 9

Number of tables: 1

Supplementary figures

Figure S1: Dried stainless steel cathode after more than 800 h of toilet wastewater electrolysis. Most of the precipitate from the bottom of the electrode had fallen off during transporting and dismantelling the electrode array.

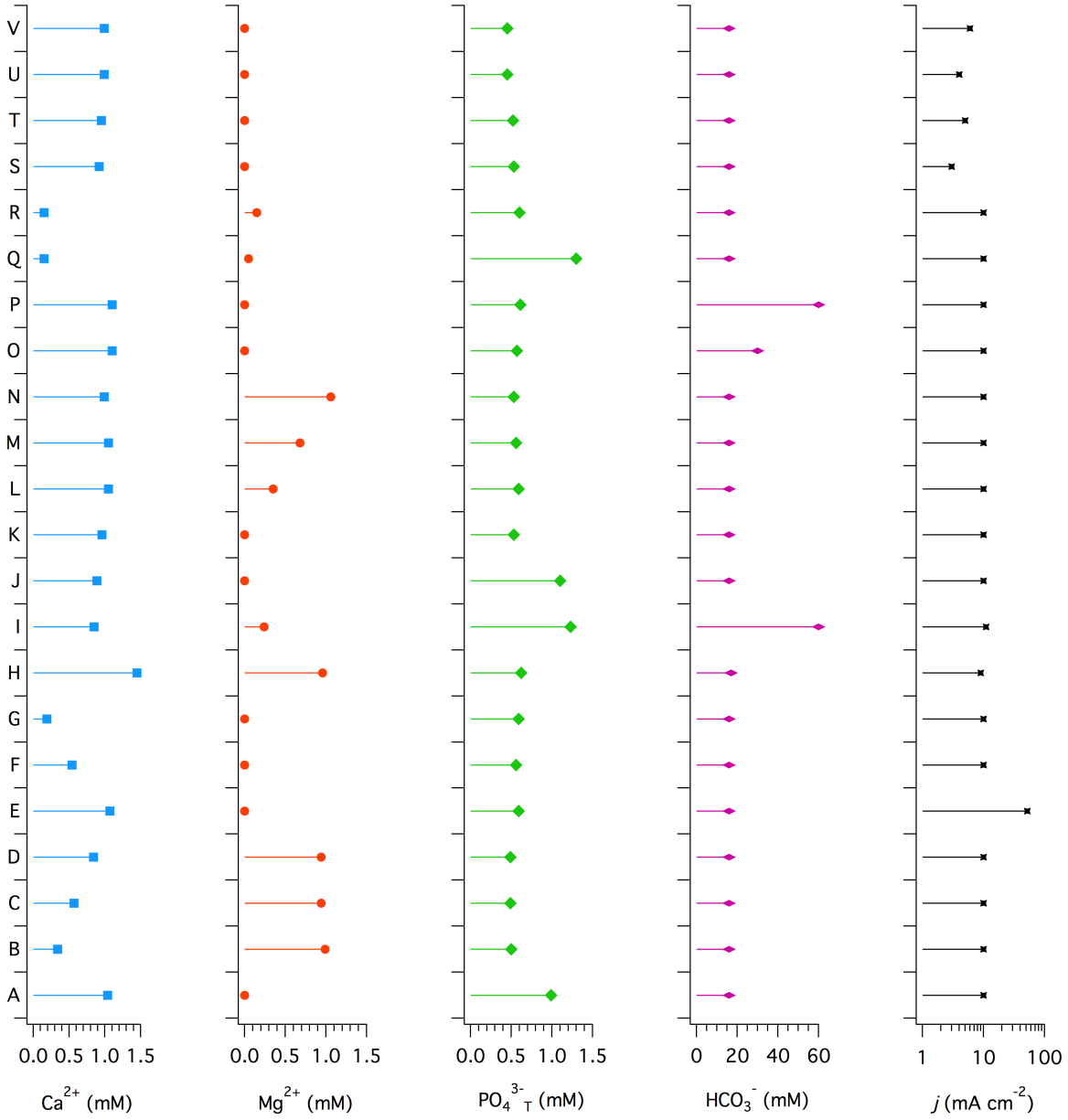
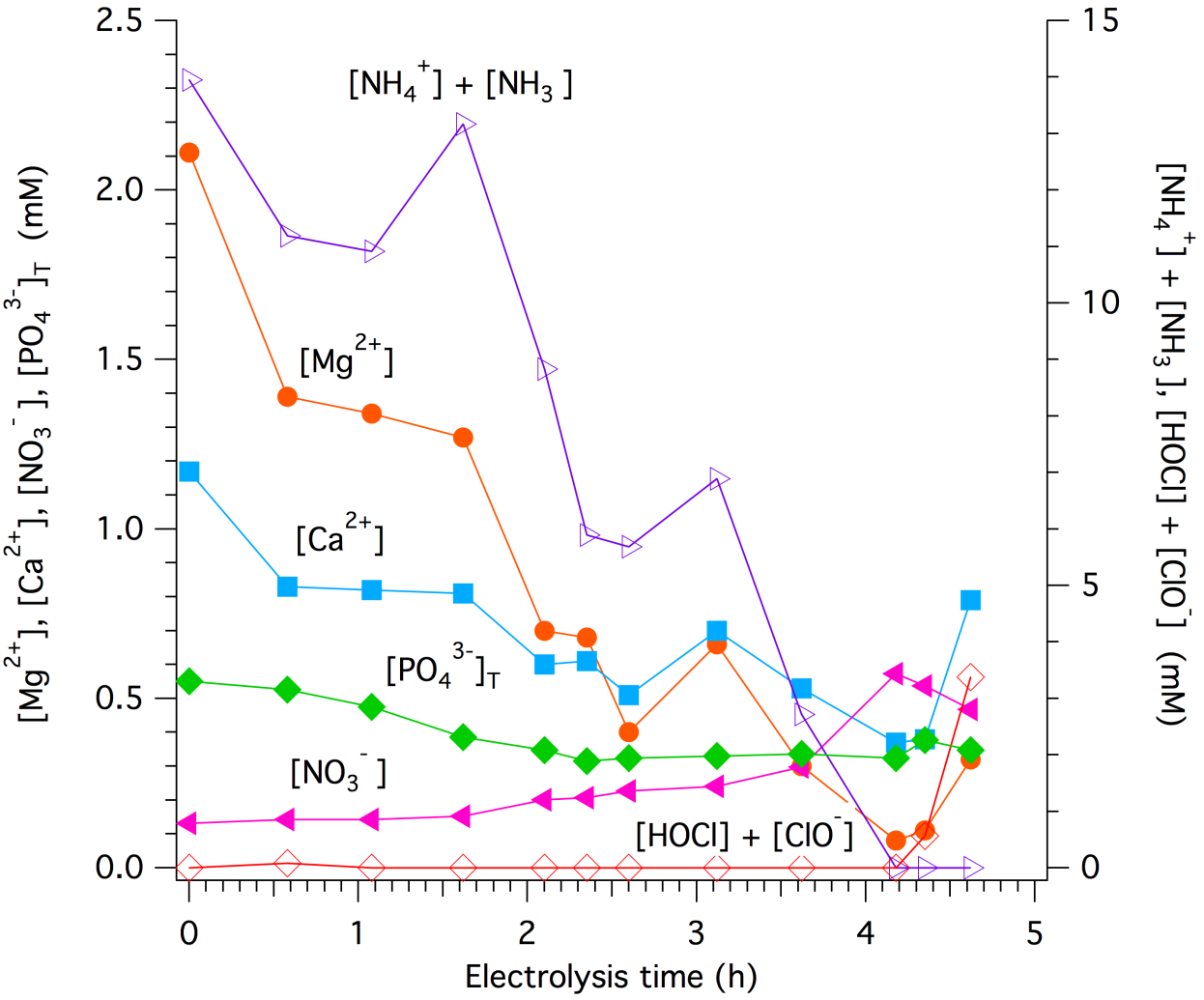


Figure S2: $[Ca^{2+}]_0$, $[Mg^{2+}]_0$, $[PO_4^{3-}]_{T,0}$, $[HCO_3^-]_0$, and current density *j* (log10 scale) for each set of triplicate experiments reported in Table S1.



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553 **Figure S3: Ammonia ($NH_4^+ + NH_3$), Mg^{2+}, Ca^{2+}, total PO_4^{3-}, NO_3^-, and free chlorine (HOCl**
554 **+ ClO^-) concentrations during electrochemical treatment (3.3 V; 50 A) of toilet**
555 **wastewater ($[Cl^-]$=80 mM) in pilot-scale reactor.**

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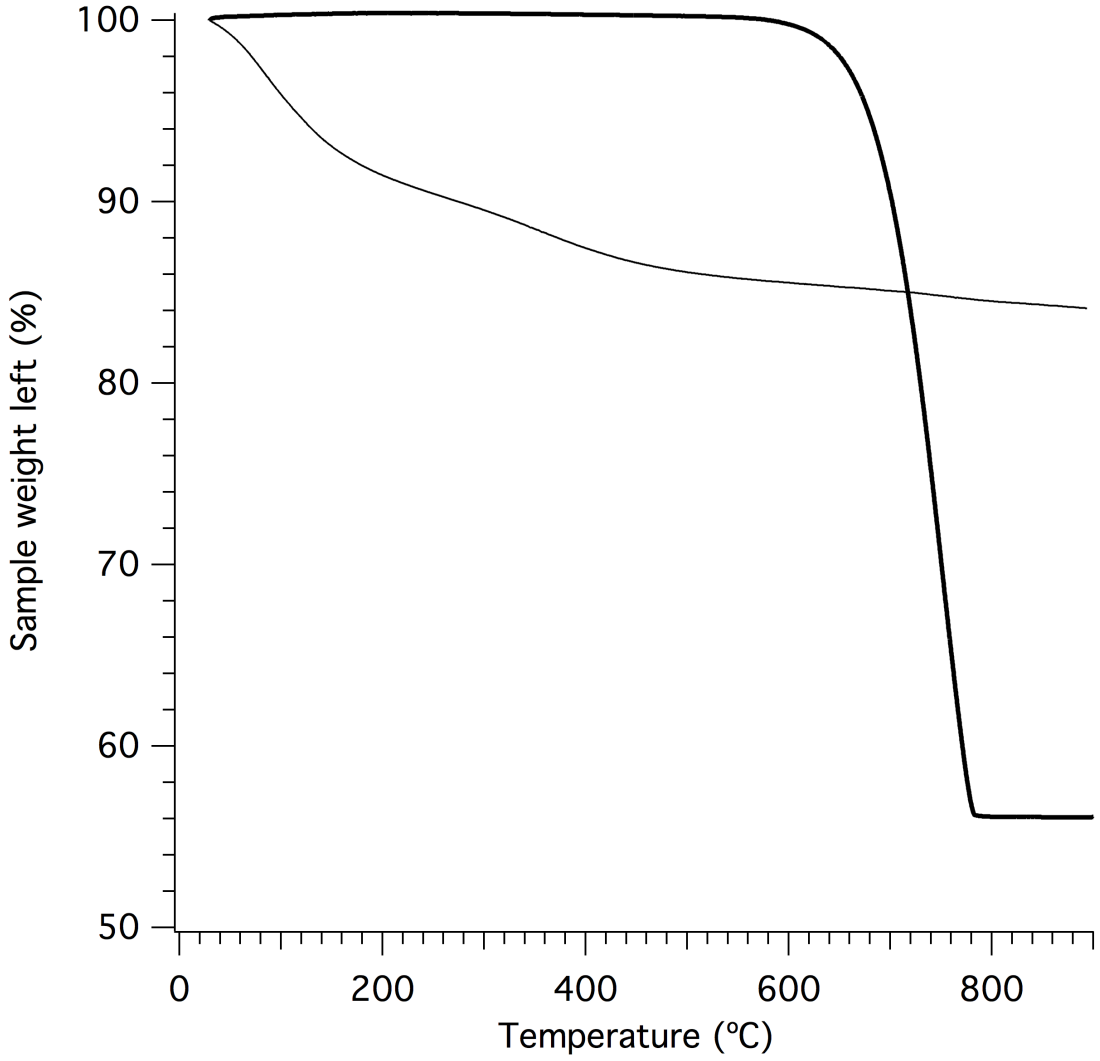

Figure S4: Thermogravimetric analysis of the precipitate collected from the electrochemical reactor (thin line) compared to calcium carbonate (thick line).

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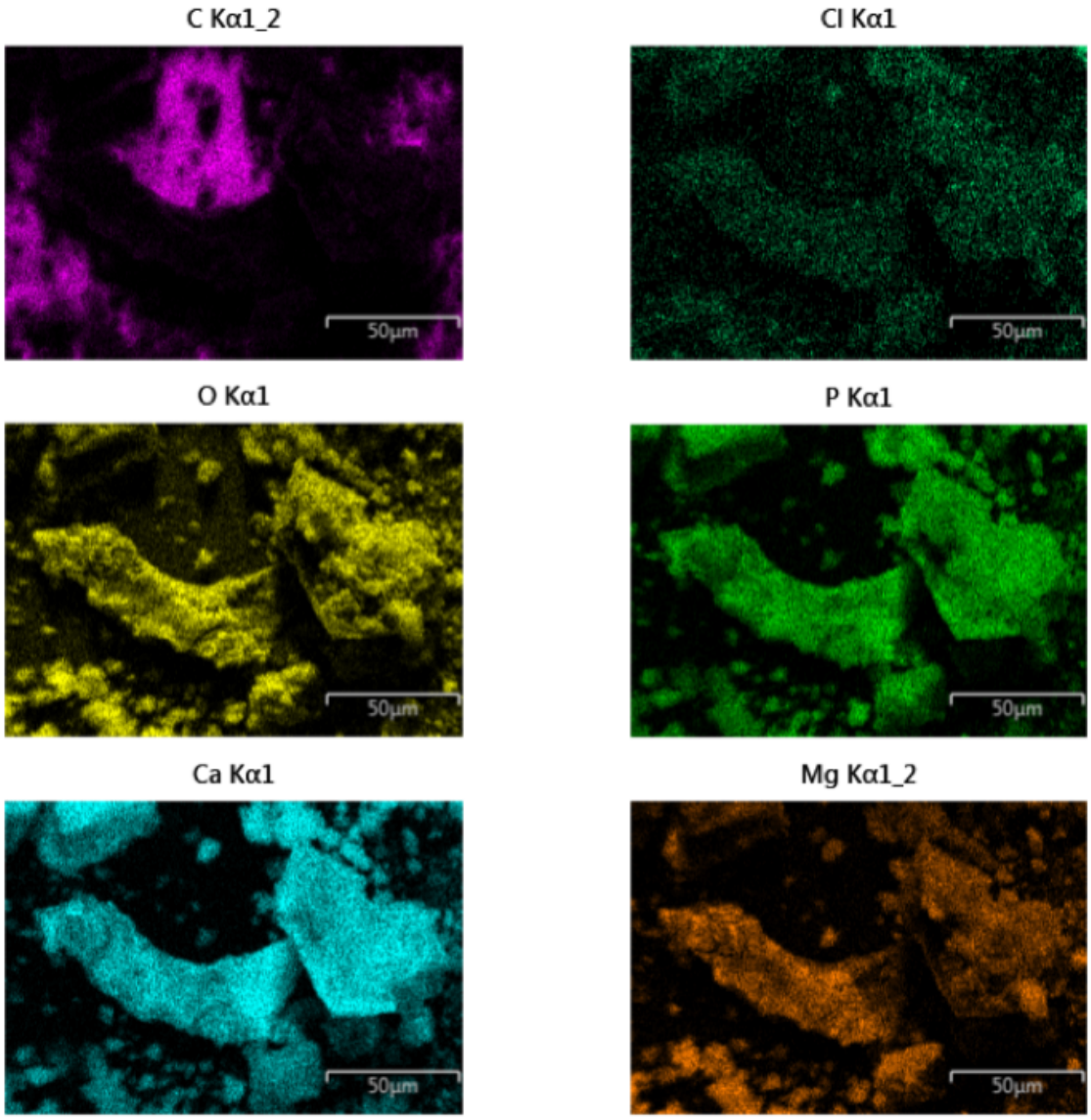

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563 **Figure S5: SEM/EDS mapping of precipitate collected from stainless steel cathodes**
564 **after several cycles of toilet wastewater electrolysis.**

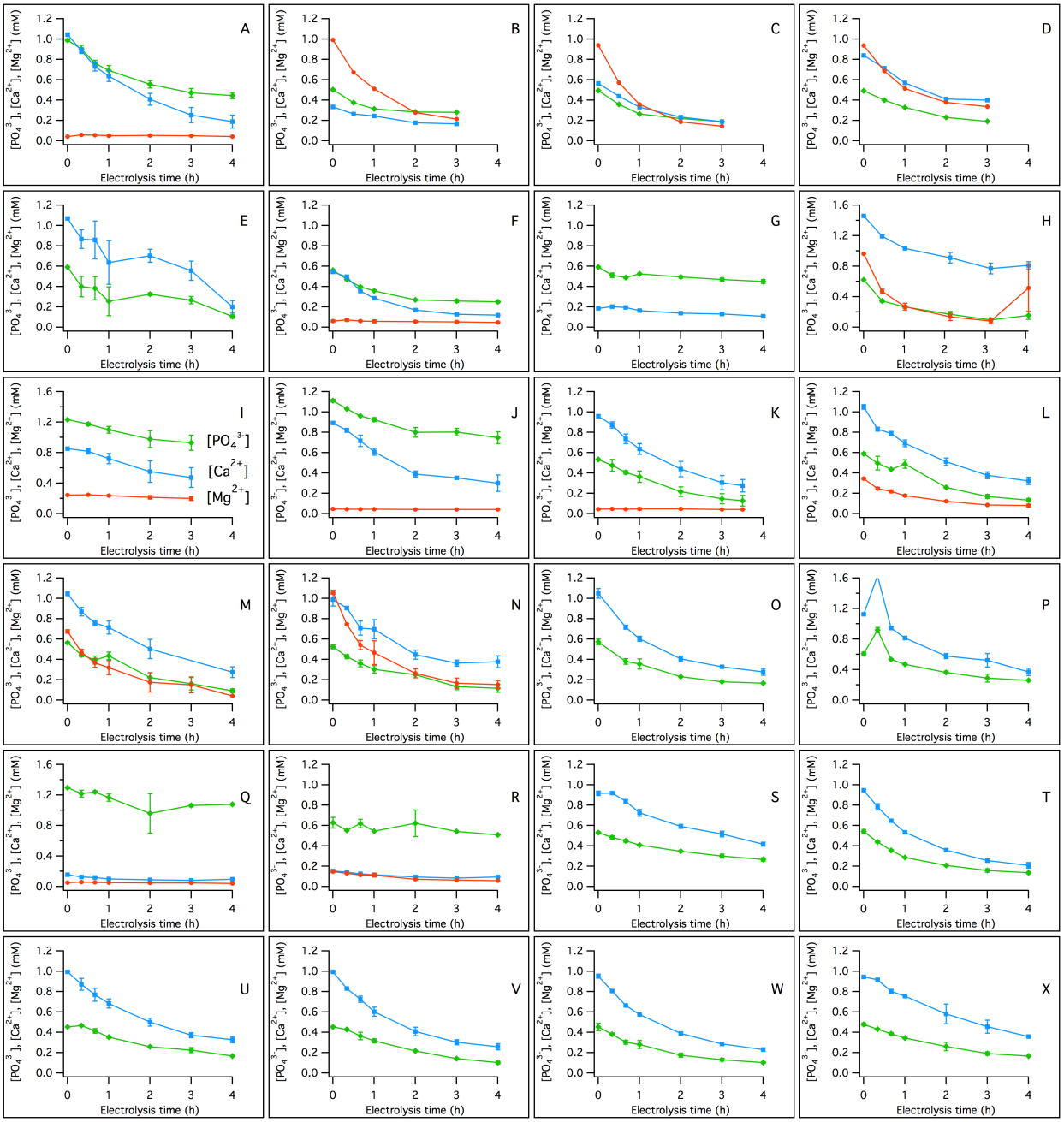

Figure S6: $[Ca^{2+}]$, $[Mg^{2+}]$, and $[PO_4^{3-}]_T$ during bench-scale synthetic wastewater electrolysis experiments. Experimental conditions for each test are detailed in Table S1. Error bars represent ± one standard deviation of 3 replicates.

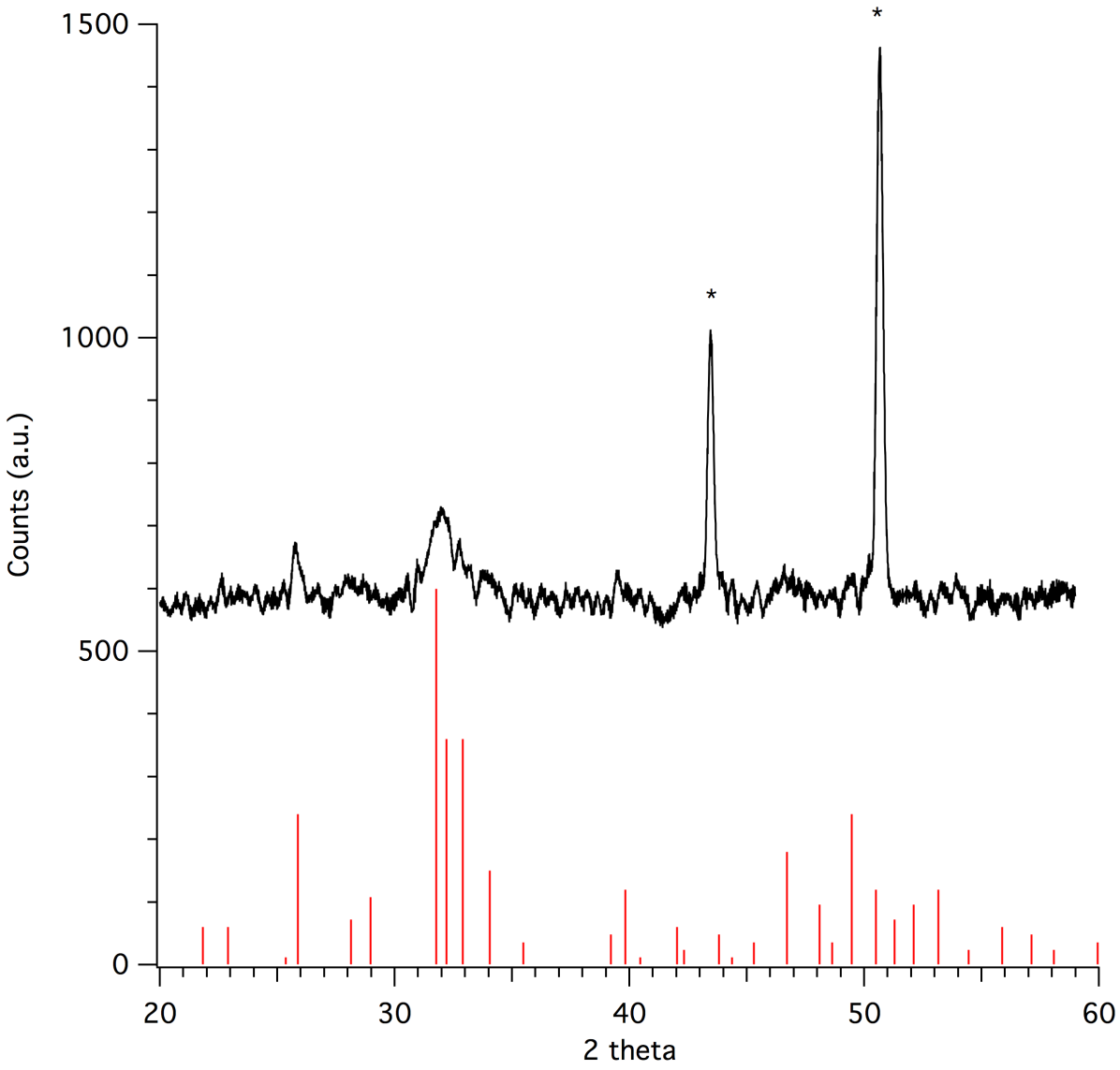

Figure S7: X-ray diffraction spectrum of a stainless steel cathode after four consecutive electrolyses of synthetic wasteater. Peaks with an asterisk are from the stainless steel. Overlaid red sticks shows pure hydroxyapatite with the highest peak normalized to 600 a.u. (ICSD# 24240 and PDF# 01-073-1731).

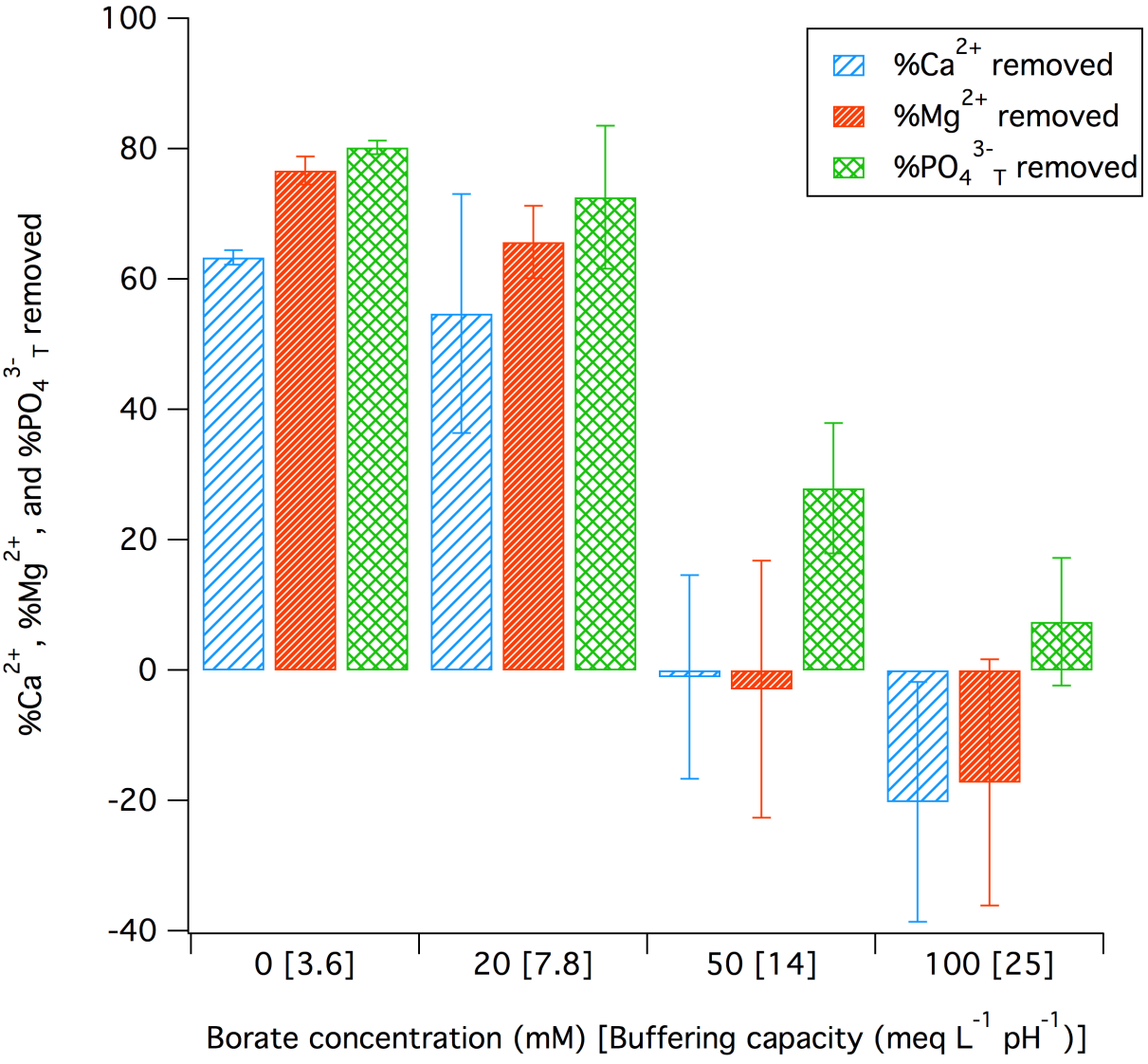

Figure S8: Percent Ca^{2+}, Mg^{2+}, and $PO_4^{3-}{}_T$ removal after potentiostatic treatment (3 h; 3.6 V; ~18 mA cm^{-2}) of synthetic wastewater buffered with sodium borate. Buffering capacities (β) of the solutions are noted in brakets. $[Ca^{2+}]_o \approx 1.0$ mM; $[Mg^{2+}]_o \approx 0.8$ mM; $[PO_4^{3-}]_o \approx 0.5$ mM; initial pH = 8.3. Error bars represent ± one standard deviation of 6 replicates.

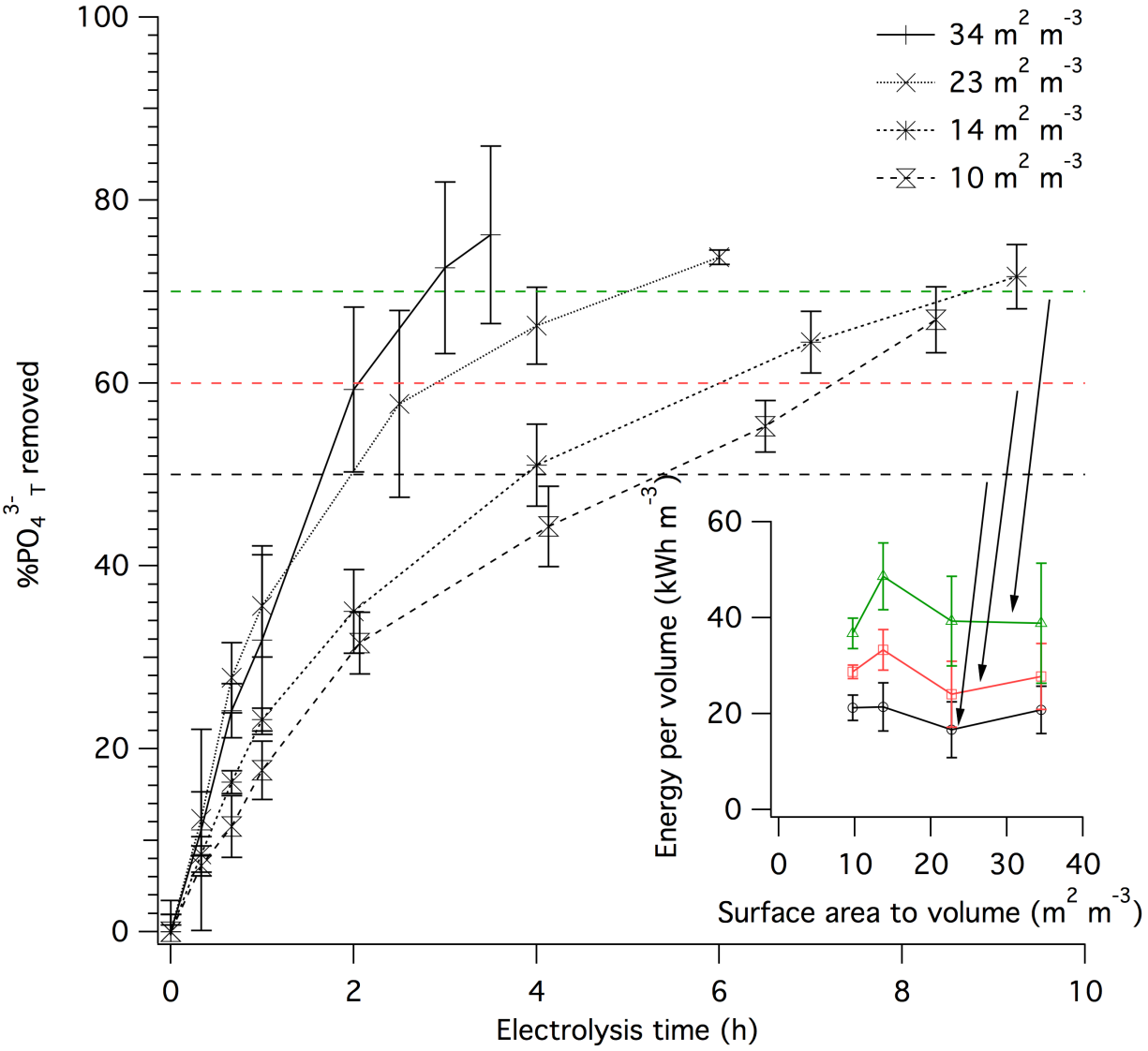

Figure S9: Percent phosphate removal during galvanostatic electrochemical treatment (10 mA cm^{-2}) of different electrode surface area to volume of synthetic wastewater ratios: 34 m^2 m^{-3}, 23 m^2 m^{-3}, 14 m^2 m^{-3}, and 10 m^2 m^{-3}. Inset: Energy per volume of wastewater required to achieve 50% (green triangles), 60% (red squares), and 70% (black circles) phosphate removal for the different volumes of synthetic wastewater. Error bars represent ± one standard deviation of 3 replicates.

Supplementary table

596 ***Table S1 – Experimental conditions for synthetic wastewater tests***

Ref #	$[Ca^{2+}]_0$ mM	$[Mg^{2+}]_0$ mM	$[PO_4^{3-}]_{T,0}$ mM	TIC_0 [a] mM	j [b] mA cm^{-2}	pH_{fin} calculated [c]	$\%Ca^{2+}$ removed	$\%PO_4^{3-}{}_T$ removed
A	1.04	0.00	0.99	16	10	9.3	82 ± 6	55 ± 3
B	0.34	0.99	0.50	16	10	9.5	50	44
C	0.57	0.94	0.49	16	10	9.6	66	62
D	0.84	0.94	0.49	16	10	9.2	52	61
E	1.07	0.00	0.59	16	52	9.7	81 ± 5.6	82 ± 2.5
F	0.54	0.00	0.56	16	10	9.7	78 ± 0.5	55 ± 2
G	0.19	0.00	0.59	16	10	9.6	41 ± 4	24 ± 4
H	1.45	0.96	0.62	17	9	9.0	47 ± 3	75 ± 8
I	0.85	0.24	1.23	60	11	8.8	53 ± 3	25 ± 8
J	0.89	0.00	1.10	16	10	8.9	66 ± 8.5	33 ± 6
K	0.96	0.00	0.53	16	10	9.5	71 ± 3	76 ± 9.5
L	1.05	0.35	0.59	16	10	9.4	69 ± 3	77 ± 3
M	1.05	0.68	0.56	16	10	9.6	74 ± 3	84 ± 4
N	0.99	1.06	0.53	16	10	9.4	62 ± 6.6	78 ± 7
O	1.10	0.00	0.57	30	10	9.6	74 ± 5	71 ± 3
P	1.10	0.00	0.61	60	10	9.4	67 ± 4	57 ± 3
Q	0.15	0.05	1.30	16	10	9.4	40 ± 3	17 ± 0.6
R	0.15	0.15	0.60	16	10	9.6	38 ± 3	19 ± 6.7
S	0.92	0.00	0.53	16	3	9.0	55 ± 1.5	50 ± 3.7
T	0.95	0.00	0.52	16	5	9.6	78 ± 2.8	75 ± 1.6
U	0.99	0.00	0.45	16	4	9.3	67 ± 3.2	63 ± 1
V	0.99	0.00	0.45	16	6	9.3	74 ± 3.1	77 ± 3

597 [a] Total Inorganic Carbon $TIC_0 = [HCO_3^-]_0 + [CO_3^{2-}]_0$. [b] current density. [c] The cathodic pH was estimated assuming that the solution at the cathode surface
598 was equilibrated (SI=1) with respect to electrochemically precipitated hydroxyapatite ($K_{sp} = 5\ 10^{-47}$) and that $[Ca^{2+}]$ and $[PO_4^{3-}]$ at the cathode were the
599 same as measured in solution at the end of the experiment (when ion concentrations had stabilized).
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