

## ***Supplementary Information***

# **Desalination of Saline Irrigation Water using Hydrophobic Metal Polymer Hydrogels**

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All reference numbers refer to references listed in the main paper

### Supplementary Information Section SA: Background Information: Metal Polymers

Metal (M) polymers fall into two principal generic groupings:

- (i)      Organo-metal polymers (e.g., patents DE60124146T2; EP1363951B1);
- (ii)     Hydroxyoxide-metal polymers [64, 65, 99,100,101].

Hydroxyoxide metal polymers can remove ions from water, during water remediation, by direct reaction, adsorption, and catalysis. The mechanisms associated with the removal of Na<sup>+</sup> and Cl<sup>-</sup> ions from water, using hydroxyoxide-metal polymers, is complex, and poorly understood.

This study focuses on hydroxyoxide metal polymers [66, 99], with the generic formulation  $[M_xO_yH_z]^{n(+/-)}_m$ . These polymers can be used to desalinate water. The base polymer is a layered double hydroxide (LDH), or a layered hydroxide salt (LHS), or a caged oligomer structure (based on FeOOH, or AlOOH)[102,103]. The dominant metal (M) is one of Fe, Ca, Mg, Mn, Al, Zn [18]. They are sometimes termed, polynuclear metal hydroxides, and have had commercial medicinal applications, as phosphate adsorbents (e.g., TWI465239B; EP2319804B1; US20130039984A1); oral iron medicines to treat anemia, incorporating a sugar (e.g., US8053470B2; CN112156109A; US20200222282A1), or a carbohydrate (e.g., US11291645B2; US11364260B2; US11478502B2).

Hydrophobic  $\text{CaCO}_3$ , has been used, to reduce the concentration of organic matter in water (JP5405454B2). A number of formulation approaches have been used, including formulation in the presence of  $\text{NaOH}$  [105].  $\text{CaCO}_3$  is hydrophilic [106], but will in the presence of  $\text{CaCl}_2$ ,  $\text{Na}_2\text{CO}_3$ , and an organic acid, form as hydrophobic particles [106]. Hydrophobic  $\text{FeOOH}$ , can be produced by, forming the LDH, in the presence of a polyacrylic acid [107]. Thin film composite membranes, incorporating hydrophilic  $\text{FeOOH}$  and polysulphone, can show high levels of water passage, combined with  $\text{Na}^+$  and  $\text{Cl}^-$  ion rejection rates of >97.5% [108]. In this study, the term hydrophobic is used (*sensu lato*) to address the situation, where  $\text{Na}^+$  and  $\text{Cl}^-$  ions are preferentially concentrated, within the porosity associated with the polymers. This allows the polymers to retain some hydrophilic characteristics, but requires capture and concentration of the ions, as either part of an Ostwald process, or a Schottky – Frenkel defect process.

### SA1. Polymer Polarity

The basic metal polymer unit is bipolar, with a negative charge associated with the  $\text{O}^{2-}$  unit, and a single positive charge, associated with the  $\text{Fe}^{3+}$  unit (Figure S1). This structuring allows  $[-\text{Fe}(\text{OH})\text{O}-]$  substitution with Pululan units, to form polymer spheres of around 0.1 micron (100 nm) diameter (DE60124146T2). The spheres form, when  $n\text{-FeOOH}$  is precipitated from  $[\text{Fe}^{n+}\text{Cl}^-]_n$  (e.g.,  $[\text{FeCl}_6]^{3-}$ ) at a pH of 7 to 12 (where the pH is controlled by the  $\text{NaOH}$  (or  $\text{KOH}$ , or  $\text{Ca}(\text{OH})_2$ ) concentration in the water (DE60124146T2; EP1363951B1)). Replacement of the  $\text{OH}^-$  group (Figure S1) with  $\text{O}^{2-}$  allows the basic unit to be tri-polar, with two negative sites, and a single positive site. This allows the simple polymer chains, to adopt the form, of a complex three dimensional network.

The presence of polar polymers, creates a situation, where polar- hydroxyoxide metal polymer spheres aggregate to form chains.

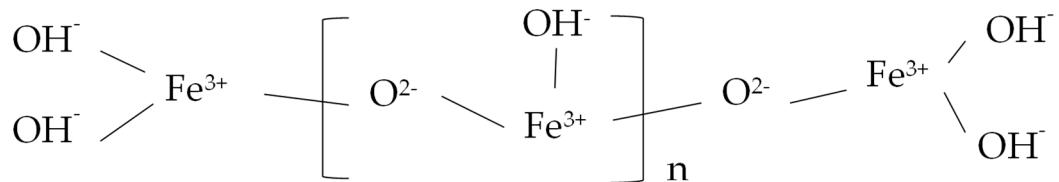


Figure S1. Schematic drawing of the basic  $\text{FeOOH}$  oligomer ( $\text{Fe}(c)$  polymer) structure.  $n$  = recurring polymer unit. The basic oligomer unit is bipolar with a positive charge and a negative charge. The remaining  $[-\text{OH}]$  group can have a neutral charge, or can be omitted, creating a positive charge, or can lose an  $\text{H}^+$  ion, creating a negative charge.

### SA2. Basic FeOOH Polymer Morphology

The base  $\text{FeOOH}$  polymer, can be described as a structure, with the generic form, illustrated in Figure S1. The initial polymers, form as, entrained spherical units, with a diameter of 1 to 20 nm (DE60124146T2; [109]). These spheres are stable, and do not agglomerate (DE60124146T2; [109]). The spheres can be supra-paramagnetic [110]. This feature, has not been specifically identified, in the polymers, created in this study (Tables S1 to S6). Aggregation of polymer spheres, may relate to an electrical polarity, or may reflect a magnetic, or paramagnetic property, associated with the spheres.

When the spheres, are formed from, ferric sulphate and urea, or are formed, in the presence of  $\text{H}_2\text{SO}_4$ , or  $\text{HSO}_4^-$ , or  $\text{SO}_4^{2-}$ , or  $\text{H}_3\text{PO}_4$ , or an organic acid, they can be supra-hydrophobic [111], or hydrophobic (JP5405454B2). The hydrophobicity can result from gas inclusions, within the sphere core, or from the formation of, hydrophobic polymers, within the sphere rim.

The initial nano-spherical units (e.g., Supplementary Information, Figure S5), can aggregate, to form larger, fluid filled, spherical units [112]. An alternative direct route, for the formation of hollow  $\text{FeOOH}$  spheres, involving a Stober process, has been identified [39, 113]. Hollow  $\text{FeOOH}$  spheres, formed using the sol-gel process, from  $\text{FeSO}_4 +$  urea, typically, have a sphere size of 500 – 1000 nm (0.5 – 1 microns) [114].

Addition of, one or more of NaOH, or KOH, or Al<sup>3+</sup>, or Mg<sup>2+</sup>, or Fe<sup>2+/3+</sup>, or Mn<sup>4+</sup>, or Zn<sup>2+</sup>, or organic salts, or HCO<sub>3</sub><sup>-</sup>, or CO<sub>3</sub><sup>2-</sup>, or OH<sup>-</sup>, or HS<sup>-</sup>, or S<sup>2-</sup>, to the sol-gel solution, can be used to functionalize the metal polymer surfaces (JP5405454B2; RU2463256C2; US9617175B2). Functionalization, requires the addition of, +ve, or -ve, sites, to the polymer surface.

It is obvious from Figure S1, that the first stage in polymer formation (and cation accretion) must be hydrophilic, via the anodic reaction,  $\text{Fe}^{2+} + 2\text{H}_2\text{O} = \text{FeOOH} + 3\text{H}^+ + \text{e}^-$  [45]. Hydrophobicity can only occur, if the rod-shaped, crystallites aggregate, and orientate, to produce a sphere. This sphere must contain an anodic surface, and a cathodic core (Figure S1). In this cathodic environment,  $2\text{H}^+ + 2\text{e}^- = \text{H}_{2(\text{g})}$  [45]. These nano-hydrogen bubbles, will adhere to the associated FeOOH crystals, by surface tension. The gas filled spheres, surrounded completely, or in part by FeOOH crystallites, will form hydrophobic, buoyant, nano-spheres.

Polymer formation, coalescence, and aggregation, is initially chaotic (involving the formation of platy or needle (lath) like nano-crystallites). It involves, an initial entrapment, of water (and associated ions). Reorganization, of the crystallites, results in, a more ordered, larger structure, containing entrapped water. This trapped water, is contained within, both dead-end pores, and the hydrated layers separating the crystallites.

Hydrophilic polymers, transport water, from the wider water body, to the dead-end pores. Hydrophobic polymers, allow water access, into the hydrated layers, separating crystallites (and can be highly hydrated). They actively sequester ions, into the dead-end pores, from the wider water body. The structure of both polymer types, is affected by the pH and Eh of the water body, together with the chemistry of the water body. Hydrophobic metal polymers, can be constructed to remove, from a feed water (US9617175B2):

1. Arsenic, including arsenate, organo-arsenates, and arsenite;
2. cyanide ions;
3. Halogen ions including chloride, bromide, iodide, and pseudohalogen ions;
4. Nitrates and nitrites;
5. Oxometal ions, including single ions, molecular clusters and colloids. These include for example: chromates ( $\text{HCrO}_4^-$ ,  $\text{Cr}_2\text{O}_7^{2-}$ ,  $\text{CrO}_4^{2-}$ ), molybdates ( $\text{MoO}_4^{2-}$ ), tungstates ( $\text{WO}_4^{2-}$ ), and vanadates ( $\text{VO}_3^{2-}$ ), associated protonated weak acid species, and particulate ions;
6. Phosphates, hydrogen phosphates, organophosphates, organophosphonates, organophosphinics, polyphosphates, including Adenosine triphosphate (ATP), Adenosine diphosphate (ADP) and Adenosine monophosphate (AMP);
7. Sulfates, bisulfates, selenates, sulfide ion,  $\text{H}_2\text{S}_{\text{aq}}$ , HS,  $\text{H}_2\text{S}$  and  $\text{S}^{2-}$ ;
8. Tellurates;
9. Cations capable of forming oxides or hydroxide ion colloids and precipitates ("oxohydroxo clusters or colloids") including  $\text{Al}^{3+}$ ,  $\text{Au}^+$ ,  $\text{Au}^{3+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Co}^{3+}$ ,  $\text{Cr}^{3+}$ ,  $\text{Cu}^+$ ,  $\text{Cu}^{2+}$ ,  $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Hg}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Mn}^{4+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Ni}^{3+}$ ,  $\text{Pb}^{2+}$ , Pu, U, Tc, Th,  $\text{Zn}^{2+}$ ,
10. Organic materials, oils and ions.

These polymers can remove chlorides (US9617175B2). They can also remove (US9617175B2) monovalent cations ( $\text{K}^+$ ,  $\text{Na}^+$ ). These polymers when treating water, form polymer concentrates, containing metal ion sulfate solutions, solid salts, solids of oxides, hydroxides, bicarbonates, and carbonates. The recovered polymers, can be used (US9617175B2) to recover a metal ore, and/or sulfate-based fertilizers, (e.g.,  $\text{K}_2\text{SO}_4$ ,  $\text{Na}_2\text{SO}_4$ ,  $(\text{NH}_4)_2\text{SO}_4$ ).

In this study, the dead-end porosity, is created within the crystallites and colloids (flocculants). It is possible to take a nano-porous substrate (e.g., porous carbon), and use a sol gel approach, to precipitate the FeOOH polymer (or equivalent), onto the surface of the substrate. The hydrophobic polymer, will then sequester the target ions, within the dead-end porosity, of the substrate (US11078093B2).

#### *SA2.1 Creating a Hydrophobic Polymer Sphere Surface*

In this study,  $\text{SO}_4^{2-}$  ions were added, to the saline feed water. Their presence, facilitates the formation of hydrophobic, or supra-hydrophobic, hollow, metal polymer spheres. Hydrophobic surfaces, operate by adsorbing, the target ion on their external surface. The ion is then passed through

the polymer to a desorption point. The effectiveness of the polymer, is a function of surface based site availability, and the ability of a polymer, to transport the ion, through the sphere, from an adsorption point, to a desorption point.

A variety of different approaches exist to create hollow hydrophobic polymer spheres. These include templating on, or functionalizing, a hollow synthetic sphere (ES2908075T3; ES2891098T3). Others, have assumed that, the metal oxyhydroxides produced by, the sol-gel approach (US20090061226A1), require the inclusion of a hydrophobic organic polymeric hydrophobic polymer (e.g., a polyolefin, a polyaromatic, a polyalkylacrylate, a polyoxirane, a polydiene, a polylactone(lactide)).

#### *SA2.2 Adsorption of Cl<sup>-</sup> ions by the spheres*

Hollow FeOOH spheres, constructed from radiating needles (0.5 micron diameter), will remove both organic dyes and heavy metals from water [36].

The hollow spheres form, through an Ostwald ripening process. This requires part of the radiating needles at the center of the sphere to dissolve, to release a Fe ion, water, hydrogen, and any ions retained in the FeOOH, prior to dissolution [37]. The addition of Na<sub>2</sub>CO<sub>3</sub> (or K<sub>2</sub>CO<sub>3</sub>) accelerates the formation of hollow nano-rod (FeOOH) particles in water containing Fe<sup>3+</sup>, Cl<sup>-</sup>, Na<sup>+</sup> and CO<sub>3</sub><sup>2-</sup> ions [38]. Simple twinning of the nano-rods, may form hollow V shaped particles. More complex twinning may form X shaped hollow particles (which may have a dumb-bell appearance)[38]. These particles will remove fluoride ions by incorporation within the polymer chain [38], e.g.,



The FeOOH can be expected to incorporate Cl<sup>-</sup> ions, within the polymer chain, as part of an anodic redox reaction, i.e.,



Where n is between 0 and 6. When n>3, this will, following subsequent dissolution, create a concentration, within the dead-end pores, of negative ions (e<sup>-</sup>).

#### *SA2.3 Adsorption of Na<sup>+</sup> ions by the spheres*

Migration of the electrons (e<sup>-</sup>), from the dead-end pores, at the center of the spheres, or in the hollow nano-rods, to the exterior of the polymer sphere, will facilitate adsorption of Na<sup>+</sup> ions. If the initial Na<sup>+</sup> redox removal reaction, is cathodic, then an adsorption reaction, may take the form [39]:



on the spheres exterior. Within the nano-sphere, or at the nano-sphere internal fluid boundary, the dissociation anodic reaction, may result in:



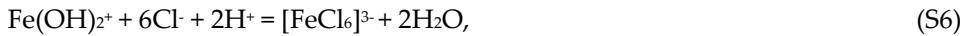
This model, assumes that the fluid, within the sphere, has a pH of <11.5, and an Eh (V), which is greater than [1.91-0.1182pH] [45]. If the pH is >11.5 and the Eh is less than [-0.675+0.591pH], the dominant dissolution Fe ion, will be HFeOO<sup>-</sup> [45]. Migration of the dissolved Fe ion, to the outer margin of the sphere, will result in the cathodic adsorption reaction, e.g.:



#### *SA2.4 Adsorption and desorption of Cl<sup>-</sup> ions*

The complex, twinned, FeOOH spicules, forming a sphere, radiate from an initial nucleus. They may all be orientated, with an anodic exterior, and a cathodic interior, or vice versa, or a combination. This orientation, will affect the ability of the sphere, to adsorb Cl<sup>-</sup> ions and Na<sup>+</sup> ions from the water.

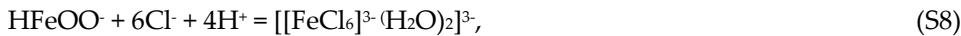
This will affect the molar selectivity for  $\text{Cl}^-$  ion removal versus  $\text{Na}^+$  ion removal. This may result in incorporation of chloride ions at the polymer's exterior, taking the form:



At the internal fluid- $\text{FeOOH}$  boundary, within the sphere, the desorption reaction may take the form:



In this example,  $\text{Cl}^-$  adsorption involves an anodic reaction, while Ostwald dissolution in the sphere center represents a cathodic reaction. Migration of the dissolved Fe ion, to the outer margin of the sphere, would result in the anodic adsorption reaction:



If the  $[[\text{FeCl}_6]^{3-}(\text{H}_2\text{O})_2]^{3-}$  unit replaces a  $[\text{OH}^-]$  unit (Figure S1), then each unit, will be able to, adsorb  $2\text{Na}^+$  or another cation, e.g.,  $2\text{Na}^+ + [[\text{FeCl}_6]^{3-}(\text{H}_2\text{O})_2]^{3-} = [[\text{FeCl}_6]^{3-}[(\text{Na}^+)_2]^{2+}(\text{H}_2\text{O})_2]^-$ . This model, implies that a molar selectivity of  $3\text{Cl}^-:\text{Na}^+$  may be expected.

#### *SA2.5 Ion Selectivity*

The adsorption-desorption model, establishes, that constructing, the sol-gel metal polymer, within the target saline water body, will create a polymer containing  $\text{FeOOH}$  spicules. These spicules will contain  $[[\text{FeOOH}]:[\text{FeCl}_n]^{(3-n)}$  and  $[[\text{FeOOH}]:[\text{Na}^+\text{FeOOH}]$ .

When the number of cathodic sites, on the sphere exterior, equals the number of anodic sites, on the sphere exterior, the spheres will be expected to remove both  $\text{Na}^+$  and  $\text{Cl}^-$  ions, in the molar ratio  $x:n$ . Selectivity is therefore a function of both the external surface charge and the rate of dissolution, at the sphere rim: sphere core fluid interface.

This Ostwald Ripening model implies that, the fluid volume within the spheres, will increase as the polymer spheres increase in size. It also implies, that the salinity of the water body, will decline, as the fluid volume, within the spheres increases.

#### *SA2.6 Working Model for Creating Adsorption Selectivity*

In this study,  $\text{R-COO}^-$ ,  $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$  ions, are added to, the feed water, to adjust polymer selectivity, associated with substitution of  $\text{OH}^-$  ions.

1. Replacement of  $[-\text{OH}]$  (Figure S1) with  $[-\text{HCO}_3]$  or  $[-\text{HSO}_4]$  is expected to reduce the number of available  $\text{Cl}^-$  removal sites  $[S_c]$ , without changing the number of available  $\text{Na}^+$  removal sites  $[S_N]$ . i.e., decrease the molar  $\text{Cl}^-:\text{Na}^+$  selectivity ratio. If the initial number  $\text{Cl}^-$  sites was  $[S_{\text{Cl}}]$ , then the expected molar ratio will be:  $[[S_{\text{Cl}}]-[S_c]]:[S_N]$ .
2. Replacement of  $[-\text{OH}]$  sites with  $[-\text{CO}_3^-]$ , or  $[-\text{SO}_4^-]$ , is expected to, reduce the number of available  $\text{Cl}^-$  removal sites, and increase the number of available  $\text{Na}^+$  removal sites, by an equivalent number. If the initial number  $\text{Cl}^-$  sites was  $[S_{\text{Cl}}]$ , then the expected molar ratio will be:  $[[S_{\text{Cl}}]-[S_c]]:[[S_N]+[S_c]]$ .

Combining  $[-\text{O}]$  sites (Figure S1) with  $\text{K}^+$ , is expected to, reduce the number of available  $\text{Na}^+$  removal sites. Combining  $[-\text{O}]$  sites with  $\text{Mg}^{2+}$ , or  $\text{Zn}^{2+}$ ,  $\text{Ca}^{2+}$ , or  $\text{Mn}^{4+}$ , or  $\text{Al}^{3+}$ , is expected to, reduce the number of available  $\text{Na}^+$  removal sites, while increasing the number of available  $\text{Cl}^-$  removal sites. Combining  $[-\text{O}]$  sites with  $\text{Mg}^{2+}$ , or  $\text{Zn}^{2+}$ , to form redox moiety structures, of the form  $[-\text{O}-\text{M}-\text{O}]$  is expected to, leave the number of, available  $\text{Na}^+$  sites, unchanged. Combining  $[-\text{O}]$  sites with  $\text{Al}^{3+}$ , to form redox moiety structures, of the form  $[-\text{O}-\text{Al}-\text{O}]_2$ , is expected to, increase the number of, available  $\text{Na}^+$  sites.

#### **SA3 Initial Ion Selectivity Model**

The  $[\text{OH}^-]$  group, within the polymer chain, (Figure S1), may control the selectivity for  $\text{Cl}^-$  and  $\text{Na}^+$  removal, from the water. Its removal by  $\text{H}^+$  ions (to form  $\text{H}_2\text{O}$ ), will create a positive charge (to allow substitution of  $\text{OH}^-$  by  $\text{Cl}^-$  and  $\text{Na}^+$  ions)[115,116,117]. The removal of the  $\text{H}^+$  creates a negatively charged vacancy. This will allow substitution of the  $\text{H}^+$  by  $\text{Na}^+$ . The polymers, when the oxidation

number is  $<3$ , form LDH (oxidation number between 2 and 3) and LHS (oxidation number = 2) structures, with aqueous inter-layers. These structures, allow for the adsorption and substitution of anions, and cations [118,119,120]. The adsorption and substitution process, is electrochemical (redox) [120] and can allow the LDH/LHS to act as membranes [40, 121,122,123].

### *SA3.1 Hydrophilic Polymers*

Mg-Al LHD have been used, to create forward osmosis (FO) membranes [123]. This observation, raised the possibility, that the hollow  $[M_xO_yH_z]^{n(+/-)}_m$  polymer spheres, could act as active osmotic desalination agents. If the polymer spheres, acted as a forward osmosis osmotic membrane, then they would be hydrophilic, and preferentially move water through the polymer, into the sphere center. This process, leaves a concentrate of  $Na^+$  and  $Cl^-$  ions, in the water body. Fresh water would be recovered, by dehydrating the recovered polymers.

### *SA3.2 Hydrophobic Polymers*

If the spheres, were supra-hydrophobic, or hydrophobic [40, 124], due to sol-gel formation associated with sulphates [111], then it may be possible, to preferentially transport  $Na^+$  and  $Cl^-$  ions, from the sphere surface, through the membrane, and into the fluid core of the sphere. This approach will result in a partially desalinated water body. The recovered spheres would, contain concentrates of the removed ions.

### *SA3.3 Supra-Hydrophobic Polymers*

Supra-hydrophobic structures, can be created [40, 124], by incorporating (into the polymer formation), one or more of ZnO ( $Zn(OH)_2$ ), MO<sub>2</sub> (e.g., TiO<sub>2</sub>, MnO<sub>2</sub>), clays (e.g., Ca-montmorillonite), feldspars (e.g., K-feldspar), polysiloxanes (-Si-O-Si- groups), carbon (n-C<sup>0</sup>, -C-C-), non-polar materials containing CH<sub>3</sub>/CH<sub>2</sub> groups, and polymers with combined chemistry.

Supra-hydrophobic structures, are characterized [40, 124] by having a rough surface, which is created through the incorporation of, one or more metals, or metal oxides, selected from the transition group metals in Periodic Table Groups III to XII (e.g., Fe, Mn, Zn). These transition metals form the polymer substrate. Their oxidation/anodization, results in the formation of, a rough supra-hydrophobic surface [40,41124,125]. These particles, can retain their supra-hydrophobic characteristics, even after contamination with, CaCO<sub>3</sub> and organic material [40,124]. It is therefore possible, to conceptualize a desalination model, where a supra-hydrophobic metal polymer sphere, is able to adsorb  $Na^+$  and  $Cl^-$  ions, on its rough polar outer surface (Figure S2). These ions, are then transported through, the polymer, to its inner fluid core (Figure S2).

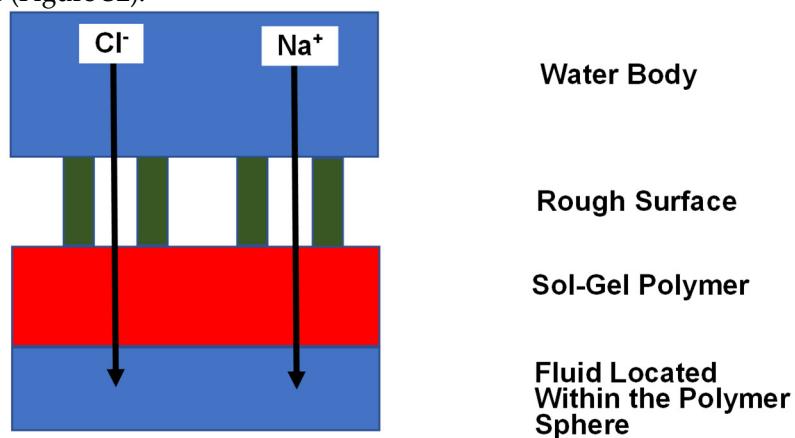


Figure S2. Conceptual desalination model using suprahydrophobic hollow metal polymer spheres.

## SA4 Working Model for Hydrophobic Adsorption

The preceding brief discussion, has indicated that alteration, of a few simple polymer formulation ingredients, may alter the adsorption selectivity, associated with a metal polymer. If the polymer is hydrophobic, or supra-hydrophobic, the surface based, adsorption sites, can be viewed as Schottky vacancies [88]. Transport through the polymer, to the fluid body, at the sphere interior, is via Frenkel defects [89] (Figure S2). Discharge of the ions, into the fluid body, results from a reversed Schottky transfer, where: (i) the discharge of  $\text{Cl}^-$  ions, is associated with, an adsorption of  $\text{OH}^-$  ions; (ii) the discharge of  $\text{Na}^+$  ions, is associated with, an adsorption of  $\text{H}^+$  ions [18]. Under this model, fluids ( $\text{H}_3\text{O}^+$ ,  $\text{H}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{OH}^-$ ) contained within the polymer sphere, are discharged over time, to the water body, and are replaced, by ions, abstracted from the water body. This process, results in a loss of buoyancy, for the entrained polymer spheres.

This model indicates, that while selectivity can be adjusted, by changing the polymer formulation, the ion removal rate constant, will be a function of, the efficiency of the ion migration associated with the Frenkel defects. This in turn, will be a function of the base metal(s) used, to formulate the polymer.

### SA4.1 Schottky and Frenkel Defects

The metal polymers contain Schottky defects [88] and Frenkel defects [89]. These defects create a mechanism and process, which allow the  $\text{Na}^+$  and  $\text{Cl}^-$  ions captured, on the outer margins, of the polymer, to be transported, to the fluid core, of the polymer sphere (Figure S2) [18], and to adjacent dead-end porosity, within the polymer complex (US10919784B2).

### SA4.2 Schottky and Frenkel Defects Operation

The Schottky and Frenkel Defect desalination model, is difficult to prove, by direct measurement. This is because, it relates to the operation of spheres, which are  $<2$  microns in diameter, and membrane thicknesses, which may be  $<0.5$  microns thick.

A potential test of the hypothesis, can be demonstrated, using brackish saline water, containing  $1.24 \text{ g Cl}^- \text{ L}^{-1} + 0.82 \text{ g Na}^+ \text{ L}^{-1}$  ( $2.00 \text{ g NaCl L}^{-1}$ ). Addition to this water of  $\text{Fe}^{n+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{Ca}^{2+}$ ,  $\text{ZnO}$ ,  $\text{MnO}_2$ ,  $+ \text{SO}_4^{2-} + \text{CO}_3^{2-}$  ions (Trial F60), would be expected to create a hydrophobic sol-gel polymer, which could remove both  $\text{Cl}^-$  ions and  $\text{Na}^+$  ions.

In this example, formulation of the polymer, in a diffusion reactor, at normal temperatures and pressures (NTP), resulted after 24 h, in a product water, containing  $0.76 \text{ g Cl}^- \text{ L}^{-1} + 0.26 \text{ g Na}^+ \text{ L}^{-1}$  ( $0.64 \text{ g NaCl L}^{-1}$ ). The Schottky and Frenkel Defect desalination model, if it occurred, would predict:

1. Evaporation of the product water, would produce a precipitate of  $\text{CaSO}_4$  crystals and some  $\text{CaCO}_3$  crystals;
2. Recovery of the polymer, followed by evaporation, of the entrained metal polymer, would breakdown the polymer spheres structures. This would result in, a release of the entrapped fluids. These would be expected to, produce a precipitate of some  $\text{CaSO}_4$  crystals, and some  $\text{CaCO}_3$  crystals. They would also be expected to show a film of tabular  $\text{NaCl}$  crystals (released from the sphere interiors) forming around the polymers.

Accordingly:

- (i) the entrained polymers were separated from the product water;
- (ii) both the product water and the recovered entrained particles were evaporated separately;
- (iii) the recovered precipitates from the evaporated samples, were examined under a microscope.

In this example:

- (i) evaporation of the product water resulted in a precipitate of  $\text{CaSO}_4$  (Figure S3a);
- (ii) evaporation of the entrained polymer precipitate (dark color) resulted in the precipitation of  $\text{CaSO}_4$ ,  $\text{CaCO}_3$  and  $\text{NaCl}$  (Figure S3b). The halite forms as a film surrounding the evaporated polymer (Figure S3c). The polymer precipitates were formed from the aggregation of polymer spheres, with an average diameter of about 1 micron (Figure S3d).

These observations, are consistent with, the abstraction model, summarized in Figure S2. They demonstrate that,  $\text{Na}^+$  and  $\text{Cl}^-$  ions are concentrated, within the sphere cores. However, the mechanism

of ion transport from the water body, could be a result of the Schottky-Frenkel conveyor mechanism, or it could be a result of the Ostwald Ripening conveyor mechanism.

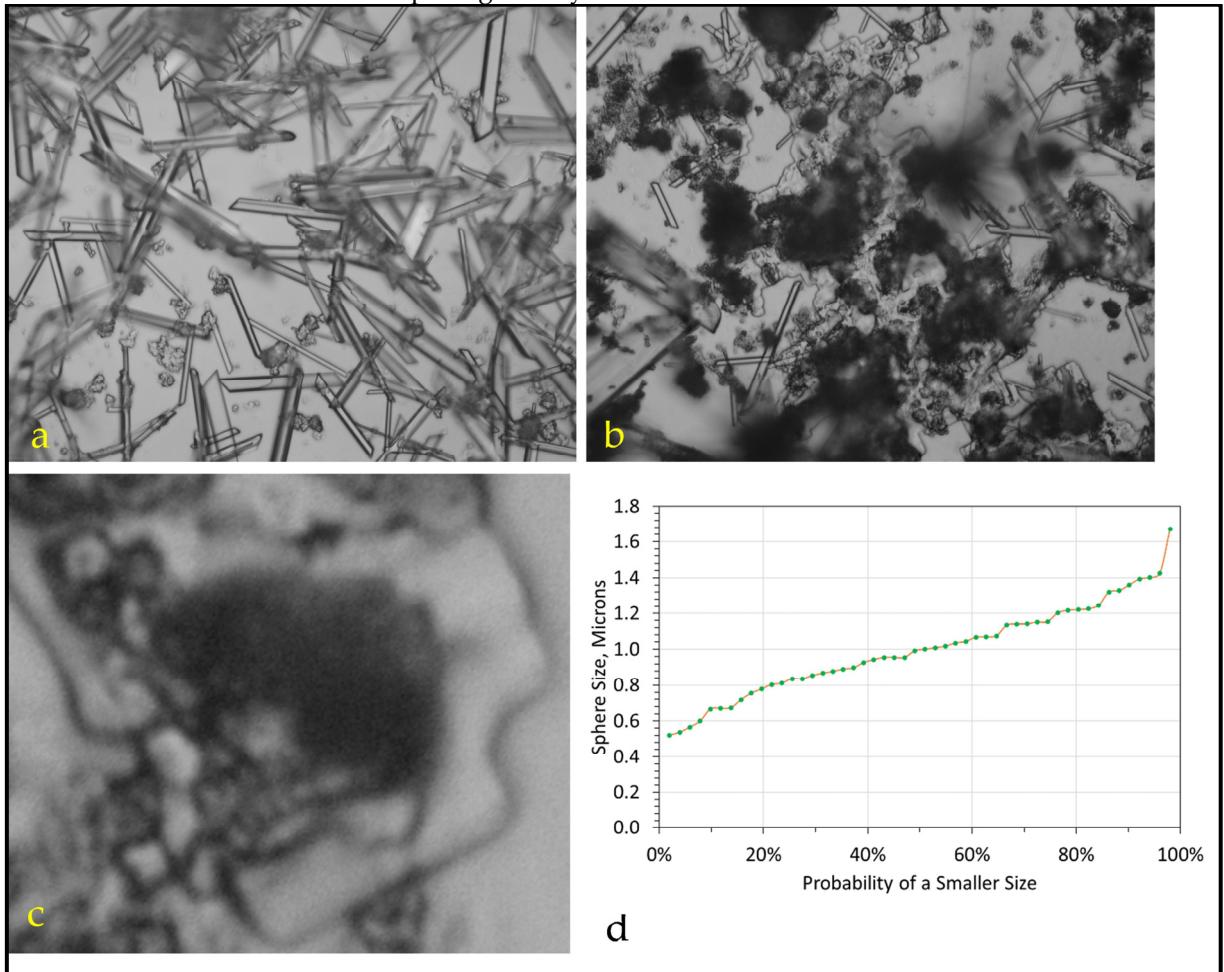


Figure S3. Trial F60. (a), Evaporated product water, showing  $\text{CaSO}_4$  (lath shaped crystals),  $\text{CaCO}_3$  (amorphous crystals); Field of view = 0.3 mm; (b), Evaporated entrained polymer particles; polymer = dark masses; lath shaped crystals =  $\text{CaSO}_4$ ; amorphous crystals =  $\text{CaCO}_3$ ; tabular sheets or films =  $\text{NaCl}$ ; Field of view = 0.3 mm. (c), Enlargement of an evaporated polymer (amorphous structure where it has broken down, and a cellular structure on two of its edges) showing a halite film forming around it. Field of view = 20 microns (0.02 mm); (d), Size distribution for the polymer spheres forming the polymer aggregates. Mean = 0.998 microns; standard deviation = 0.255; number of measurements = 50.

### SA5 Janus Characteristics

When the polymers are entrained, as nano-particles in water (Figure S3d; S4), they operate as motile Janus particles [126,127]. The Janus particles, tend to form as entrained multi-polar spheres (formed from aggregated oligomers), which encapsulate a central fluid core. The Janus particle spheres, may contain hydrogen gas, within their fluid core, to provide a degree of buoyancy. This fluid, is a mixture of water, hydrogen (and other desorbed gases), and abstracted ions. The Schottky defects and Frenkel defects are used (Figure S2), to migrate the  $\text{Na}^+$  and  $\text{Cl}^-$  ions, (which are adsorbed from the water body, at the sphere-water interface), through the polymer, and into (i) the encapsulated fluid body, within the sphere, and (ii) adjacent inter-structural dead-end porosity within M(c) polymers. This change, results in an increase in fluid density, combined with a loss of buoyancy, resulting in aggregation and agglomeration of particles. This is combined with a loss of mobility (Figure S3b,S3c).

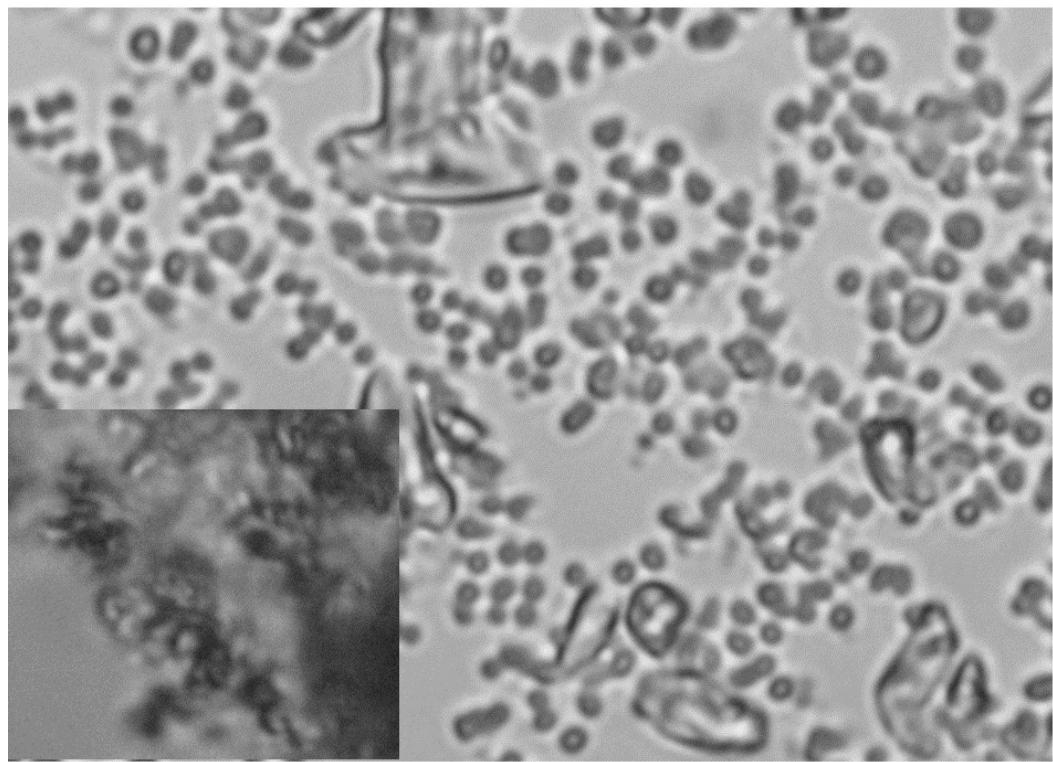


Figure S4. Trial F21: Product water showing separate colloidal polymer spheres, 1 – 1.5 microns in diameter (Field of view = 63 microns); The larger particles are simple polymer sphere aggregates. Inset is aggregated colloidal polymer spheres, 2.8 – 3.0 microns in diameter (Field of view = 36.7 microns). Cl<sup>-</sup> ion removal = 1.1%; Na<sup>+</sup> ion removal = 62.4%; Feed water contained: Cl<sup>-</sup> = 25.47 g L<sup>-1</sup>; Na<sup>+</sup> = 23.95 g L<sup>-1</sup>.

## SA6 Polymer Terminology

In this study, the relationship indicator [:] is used here to signify chemical bonding, e.g., n-Fe<sup>0</sup>:Fe(b). The relationship indicator [@] is used here to either signify physical bonding, e.g., n-Fe<sup>0</sup>@n-C<sup>0</sup>, or adhesion bonding, or an undefined bonding relationship. The following terminology approach is used in this study [64,65,66, 128]:

1. M(a) = mono-nuclear hydroxyl complexes, e.g., Fe(OH)<sub>2</sub> (molecule), Fe(OH)<sub>3</sub> (molecule), Fe(OH)<sub>4</sub><sup>-</sup>, Fe<sub>2</sub>(OH)<sub>2</sub><sup>4+</sup> and Fe<sub>3</sub>(OH)<sub>4</sub><sup>5+</sup>, some small polymers, and instantaneous reaction products.
2. M(b) = polynuclear hydroxyl complexes, with medium and high molecular mass, e.g., Fe(OH)<sub>2</sub><sup>4+</sup>; Fe(H<sub>2</sub>O)<sub>6</sub><sup>3+</sup>; Fe(O<sub>2</sub>H<sub>6</sub>)<sup>3+</sup>; Fe<sub>3</sub>(OH)<sub>4</sub><sup>5+</sup>; Fe<sub>5</sub>(OH)<sub>6</sub><sup>6+</sup>, etc. e.g., CN113880208A; CN210261456U; CN210261453U; CN210261454U; CN113197151A;
3. M(c) = larger polymer or colloidal species, which include inert macro-molecular coagulation, of the form M(OH)<sub>x</sub> and MOOH.

Most metal polymers are a mixture of M(a) + M(b) + M(c). M(a) tend to dominate acidic waters while M(b) and M(c) tend to dominate neutral and basic waters [129]. High molecular weight organics and heavy metals tend to be adsorbed/removed by M(b). M(c) polymers are effective at removing lower molecular weight organics [129].

The target polymer is M(a) and M(b), but can include SO<sub>x</sub>, CO<sub>x</sub>, C<sub>x</sub>H<sub>y</sub>, and C<sub>x</sub>H<sub>y</sub>O<sub>z</sub> species. The polymers can be unsupported (e.g., n-Fe(b)), or supported (e.g., SiO<sub>2</sub>@Fe(b); Fe<sup>0</sup>@Fe(b); Fe<sup>0</sup>:Fe(b)@C<sup>0</sup>; C<sup>0</sup>@Fe(b)). Metal polymers can have a positive charge, or a negative charge. Aggregated polymer colloids, are commonly polar, or multipolar, and aggregate to form, ribbons, chains and nets of polymer. In this study, the term ZVI desalination, is used to refer to desalination using metal polymers.

Items, added to, the sol-gel polymer formulation ingredients, which are designed to increase roughness (Figure S2), to create a polymer with supra-hydrophobic characteristics, are shown using the relationship indicator [@].

## SA7 Supra-hydrophobic and Hydrophobic Metal Polymers

The metal polymers, which have been found, to act as desalination agents include:

1. n-Fe<sup>0</sup>:Fe(b): This polymer is used in a continuously stirred environment (US10919784B2; [131,132,133]), or fluidised bed environment (ES2598032; US9624113B2; US9828258B2; US2018/0009678A; [133]), or a fluidised bubble column environment (US10919784B2), or a diffusion environment (GB2520775A). This polymer removes Na<sup>+</sup> and Cl<sup>-</sup> ions from the water over a period of <1 hr to 300 days. Ion removal with time, follows either a pseudo-first-order reaction pattern, or a pseudo-second-order reaction pattern. Preferential selectivity for Cl<sup>-</sup> removal relative to Na<sup>+</sup> removal was demonstrated in the presence of HCO<sub>3</sub><sup>-</sup>, where the Na<sup>+</sup> ions are retained as NaHCO<sub>3</sub>.
2. m-Fe<sup>0</sup>:Fe(b)@C<sup>0</sup>: This polymer (GB2520775A), when placed as pellets in saline water, removes Na<sup>+</sup> and Cl<sup>-</sup> ions from the water and sequesters them in dead-end porosity, within the polymer. Ion removal with time, follows a pseudo-zero-order reaction pattern. Desalination ceases after 3000 – 5000 hours.
3. Fe<sup>0</sup>:Fe(b)@Urea: This polymer, when used in a rapid catalytic pressure swing adsorption desorption(RCPsAD) reactor (GB2470764B), containing oxygenated saline water, catalyses the formation of entrained n-Fe(a,b,c) polymer colloids, which scavenge and sequester Na<sup>+</sup> and Cl<sup>-</sup> ions from the water. Desalination ceases after 0.5 - 3 hours. Reaction order: Not determined. Observed ion removal selectivity: Feed water (17 m<sup>3</sup>) contained 2.18 Moles Cl<sup>-</sup>: 1 Mole Na<sup>+</sup>. Average Cl<sup>-</sup> molar removal = 32.1%; Average Na<sup>+</sup> molar removal = 28.5%. Average feed water salinity = 4.73 g Cl<sup>-</sup> L<sup>-1</sup>; 2.17 g Na<sup>+</sup> L<sup>-1</sup>.
4. SiO<sub>2</sub>@Fe(b)@Urea: This polymer, when used in a rapid catalytic pressure swing adsorption desorption reactor (GB2470764B), containing oxygenated saline water, catalyses the formation of entrained n-Fe(a,b,c) polymer colloids, which scavenge and sequester Na<sup>+</sup> and Cl<sup>-</sup> ions from the water. Desalination ceases after 1 - 50 hours. Reaction order: Not determined. Observed ion removal selectivity: Feed water (43 m<sup>3</sup>) contained 1.34 Moles Cl<sup>-</sup>: 1 Mole Na<sup>+</sup>. Average Cl<sup>-</sup> molar removal = 47.4%; Average Na<sup>+</sup> molar removal = 44.1%. Average feed water salinity = 2.53 g Cl<sup>-</sup> L<sup>-1</sup>; 1.67 g Na<sup>+</sup> L<sup>-1</sup>.
5. Sol-gel n-M(a,b,c) polymers [18]: This polymer grouping, can be self-assembly, within the saline water. Ion removal with time, commonly follows a pseudo-first-order reaction pattern, when used in a diffusion environment. Desalination ceases after 0.1 – 1000 hours, depending on the water composition and polymer used. The sol-gel approach, uses the sol-gel, to remove Na<sup>+</sup> and Cl<sup>-</sup> ions from the water, leaving a residual partially desalinated water product (Figure S6). Ion removal selectivity, is a function of the sol-gel ingredients used, and the water composition.

Sol-gel procedures can produce M(a,b,c) polymers, but when the ingredients contain a Fe salt, they can form magnetic n-Fe<sub>3</sub>O<sub>4</sub>. Functionalised magnetic n-Fe<sub>3</sub>O<sub>4</sub>, has been used to directly desalinate water, by ion adsorption. These magnetic n-Fe<sub>3</sub>O<sub>4</sub> particles, have also been incorporated, within forward osmosis desalination membranes [134,135].

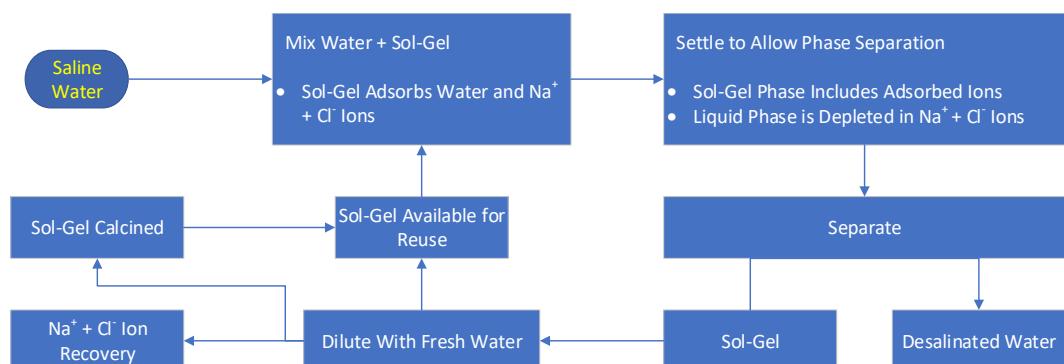
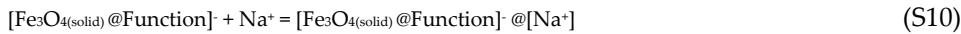
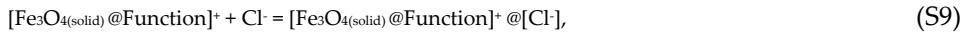


Figure S5. Process flow diagram for the sol-gel ZVI desalination process

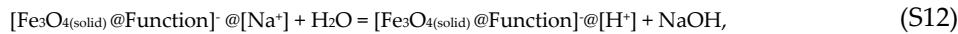
## SA8 Polymer Functionalization

All five polymer groupings, have been demonstrated to reduce water salinity, in surface based reactors (diffusion reactors, fixed bed reactors, traveling line reactors and RCPSAD reactors).

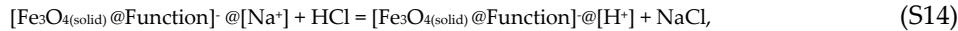
Selectivity for ion removal, is a function of, the degree of functionalisation present. Placement of Fe<sup>0</sup> in water, will initially result in a functionalisation of the form: 2Fe<sup>0</sup> + H<sub>2</sub>O = Fe@OH<sup>-</sup> + Fe@H<sup>+</sup>. This concept has been used, to reduce water salinity, using n-Fe<sup>0</sup>, with a contact time of <10 minutes, where: 2Fe<sup>0</sup> + H<sub>2</sub>O + Na<sup>+</sup> + Cl<sup>-</sup> = Fe@OH<sup>-</sup> + Fe@H<sup>+</sup> + Na<sup>+</sup> + Cl<sup>-</sup> = Fe@OH<sup>-</sup>@Na<sup>+</sup> + Fe@H<sup>+</sup>@Cl<sup>-</sup> (e.g., ES2598032). The n-Fe<sup>0</sup> can be functionalised, by the adsorption of cationic sites, or anionic sites, onto its surface, or the n-Fe<sup>0</sup> can be precipitated onto (or into), a functionalised support [136,137,138,139,140,141]. This approach has been used to provide functionalised Fe<sub>3</sub>O<sub>4</sub> particles to desalinate water (e.g., US8636906B2). There are a variety of adsorbents, which can be used to functionalise Fe<sup>0</sup>, Fe<sub>3</sub>O<sub>4</sub>, or another metal polymer particle type [142,143,144,145,146,147]. They operate by adsorbing Na<sup>+</sup> and Cl<sup>-</sup> ions, e.g.,



For ease of representation, the base polymer unit shown in the Equations is Fe<sub>3</sub>O<sub>4</sub>. The choice of cationic, or anionic, functionalisation, allows the particle, to selectively remove either Na<sup>+</sup> or Cl<sup>-</sup> ions. A typical Fe<sup>0</sup>:Fe(b), or Fe<sup>0</sup>:Fe(b)@C<sup>0</sup> polymer, will contain a similar proportion of cationic and anionic sites. This allows the removal of Cl<sup>-</sup> and Na<sup>+</sup> ions, in roughly equal molar proportions. The adsorbed Na<sup>+</sup> and Cl<sup>-</sup> ions, can be removed by washing the recovered polymer in water [[H<sup>+</sup>][OH<sup>-</sup>]], to produce a product water containing NaCl, e.g.,



A more focused Na<sup>+</sup> and Cl<sup>-</sup> ion recovery process uses a two stage approach, e.g.,



The formation of sol-gel M(a,b,c) polymers, will allow selective functionalisation to occur. This can result from the specific charge on the n-M(a,b,c) polymer, or the addition of a competitor ion to the water, which preferentially attracts one of Na<sup>+</sup> and Cl<sup>-</sup> ions.

## SA9 Solvent Based Desalination

The solvent-based desalination (SBD), approach [148,149,150,151] uses a solvent to adsorb water (for future recovery from the solute (saline water)), leaving a residual brine product (Figure S6). Two variants of the SBD approach (e.g., patents US3088909; US3177139; US 3408290; US8119007B2 US2022/0250941A1) have been defined. They are:

1. SBD1: Solvent driven water extraction (SDWE) [151], where the solvent is hydroscopic (hydrophilic), and preferentially adsorbs water, leaving a concentrated brine waste product (e.g., US8119007B2; US8501007). Suitable solvents include, but are not limited to: alcohols, aliphatic acids (R-COOH, where R is a nonaromatic (aliphatic) hydrocarbon, e.g., ethanoic, propionic, etc.), amines; epoxide-based polymers, ethers, and ionic liquids. They possess hydrophilic moieties that allow for selective solvation of water from the residual brine.
2. SBD2: Solvent driven fractional crystallization (SDFC) [151], selectively uses solvents to preferentially selectively remove target species, by precipitation, from a brine, e.g., NaCl, KCl, LiCl, etc.

The polymer model developed in this study, treats the solid metal polymer (within the spheres) as a hydrophobic solid solution. This solid solution is a “restricted solid solution”, as its components only completely dissolve (and form aqueous ions) under specific redox (Eh and pH) conditions. Within a solid solution, one group of components acts a solvent, while the other group of components acts as a solute.

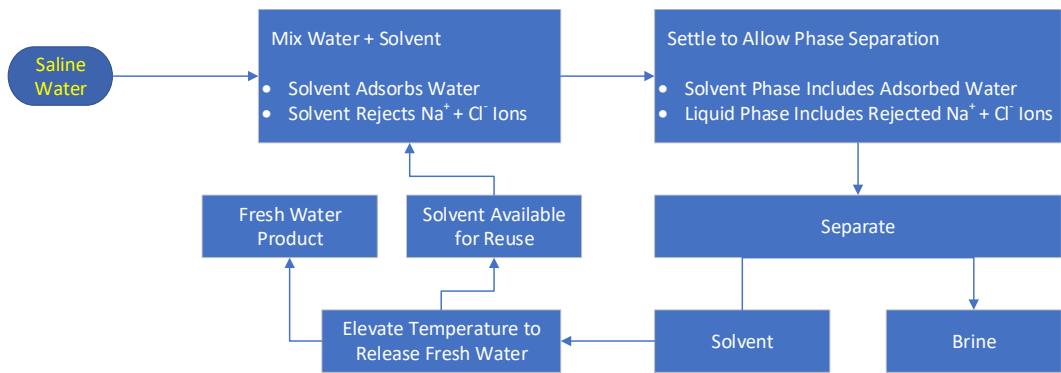


Figure S6. Process flow diagram for the solvent-based desalination process

### SA10 Crown Ether Based Desalination

The crown ether desalination approach (US2011/0147314A1), creates a desalination sponge, using crown ethers placed on a scaffold (e.g., nano-porous, high surface area, active carbon) [26,27]. The active carbon scaffold, is attached to an electrode (US2011/0147314A1). Crown ether type moieties, are attached to the scaffold (Figure S7a). Crown ethers, are heterocyclic chemical compounds, that consist of a ring containing several ether groups ((-R-O-), e.g., cyclic oligomers of ethylene oxide, where the repeating unit is -CH<sub>2</sub>CH<sub>2</sub>O- (US2011/0147314A1)). This cyclic structure, creates a central vacancy, for a monovalent cation. Crown ethers have the terminology [M-Crown-N], where M = the number of carbon + oxygen atoms in the ring; N = the number of oxygen atoms in the ring. 12-Crown-4, preferentially remove Li<sup>+</sup> ions; 15-Crown-5, preferentially remove Na<sup>+</sup> ions. The azo version replaces an oxygen with nitrogen; 18-Crown-6, preferentially remove K<sup>+</sup> ions; 21-Crown-7, preferentially remove Cs<sup>+</sup> ions ((US2011/0147314A1; [28,29,30,31,32]).

Placement of the electrodes in saline water (with no charge), will result in a redox physical adsorption of Na<sup>+</sup> ions, into the central part of the crown (Figure S7b). This creates a positive charge on the crown. This charge results in a spectator concentration of the Cl<sup>-</sup> ions to the crown exterior, within its hydration shell (Figure S7b). This simple process, has been demonstrated (using an uncharged aza-15-crown-5 ether (C<sub>10</sub>H<sub>21</sub>NO<sub>4</sub>; molecular weight = 219.28 g Mole<sup>-1</sup> (US2011/0147314A1)), to reduce the saline water salinity, from 33 g L<sup>-1</sup> to 5 g L<sup>-1</sup>.

Desalination of 10 m<sup>3</sup> of flowing saline water, will require the removal of 4791 moles of NaCl. This removal, without recycle (or regeneration) of the crown ether, will require at least 4791 moles (and probably > 8000 moles) of C<sub>10</sub>H<sub>21</sub>NO<sub>4</sub>. US2011/0147314A1 has established, that once the required salinity is achieved, the adsorbed Na<sup>+</sup> and Cl<sup>-</sup> ions, can be desorbed from the tethered crown. This approach used is to:

1. Cease the water flow, leaving the electrodes (Figure S7) immersed in oxygenated water;
2. Place a DC current across the electrodes, where the tethered crown is attached to the anode. This has the effect of placing a positive charge on the tethered crown. The net effect is to expel the Na<sup>+</sup> ions into the water body, to form an NaOH solution. The spectator Cl<sup>-</sup> ions become tethered to the anode (Figure S7c).
3. Replacing the water, and reversing the current will result in the tethered crown being attached to the cathode. This has the effect of placing a negative charge on the tethered crown. The net effect is to expel the Cl<sup>-</sup> ions into the water body (Figure S7d).

This tethered crown process, can be used to preferentially extract and concentrate Li<sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, and Cs<sup>+</sup> ions, as MOH<sub>aq</sub> species. It can also be used, to preferentially remove some divalent ions (e.g., Mg<sup>2+</sup>

[33]). This adsorption principal, is similar to the adsorption, associated with metal  $\text{FeO}_x\text{H}_y$  polymers. Constructing a metal polymer as  $\text{C}^0@\text{n-Fe(a,b,c)}$ , or  $\text{Fe}^0:\text{n-Fe(a,b,c)}$ , where  $\text{C}^0$  (or  $\text{Fe}^0$ ) is an electrode, and  $\text{Fe(a,b,c)}$  replaces the crown ether structure (Figure S7), will allow passive adsorption of a tethered  $\text{Cl}^-$  ion, with an accompanying spectator  $\text{Na}^+$  ion. Application of a DC current, will then allow preferential, and selective, desorption of  $\text{Na}^+$  ions and  $\text{Cl}^-$  ions (e.g., Figure S7). This latter approach, offers an alternative route, to the crown ether approach, and may allow the  $\text{Na}^+$  and  $\text{Cl}^-$  ions, extracted by the hydroxyoxide metal polymers, to be selectively recovered, following desalination.

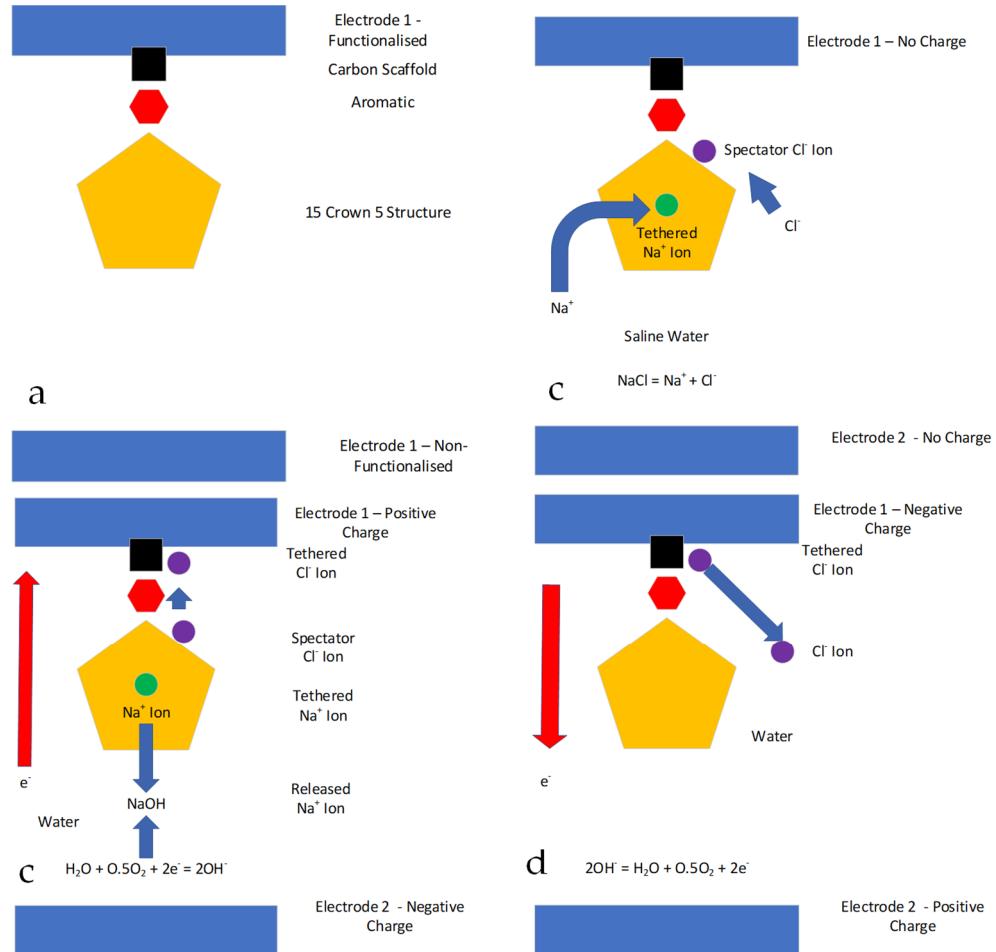


Figure S7. Schematic representation of the Tethered Crown desalination process. (a), Initial structure; (b), Operation in flowing saline water, providing removal of  $\text{Na}^+$  and  $\text{Cl}^-$  ions; (c), Regeneration, to preferentially remove  $\text{Na}^+$  ions and tether  $\text{Cl}^-$  ions; (d), Regeneration to restore the Tethered Crown to its initial position by releasing the adsorbed  $\text{Cl}^-$  ions into the water.

Creating a polymer environment, where  $\text{FeOOH}$  is produced from  $\text{Fe}^{2+}$  ions, creates a charged polymer (i.e., a capacitor (Figure S8)). Electrochemical charging is associated with an increase in oxidation number, while an electrochemical discharge is associated with a decrease in oxidation number. Placement of  $\text{MnO}_2$  into an environment, where  $\text{MnO}_2$  breaks down to form  $\text{MnOOH}$  and then  $\text{Mn}(\text{OH})_2$ , is a discharge reaction (Figure S8). Once the charged ( $\text{MOOH}$ ) polymer has formed, the charge contrast across the polymer is controlled by the oxidation number of hydrogen. It is present as  $\text{H}^+$  (oxidation number = 1) in the water body and initially as  $\text{H}_2$  (oxidation number = 0), in the fluid contained within the polymer dead-end pores. This maintains an anodic  $\text{MOOH}$ :water body interface and a cathodic  $\text{MOOH}$ :dead-end

pore interface. Placement of  $\text{MnO}_2$  in the water (Figure S8) is expected to act as a cathodic center, relative to  $\text{FeOOH}$  and  $\text{MnOOH}$ .

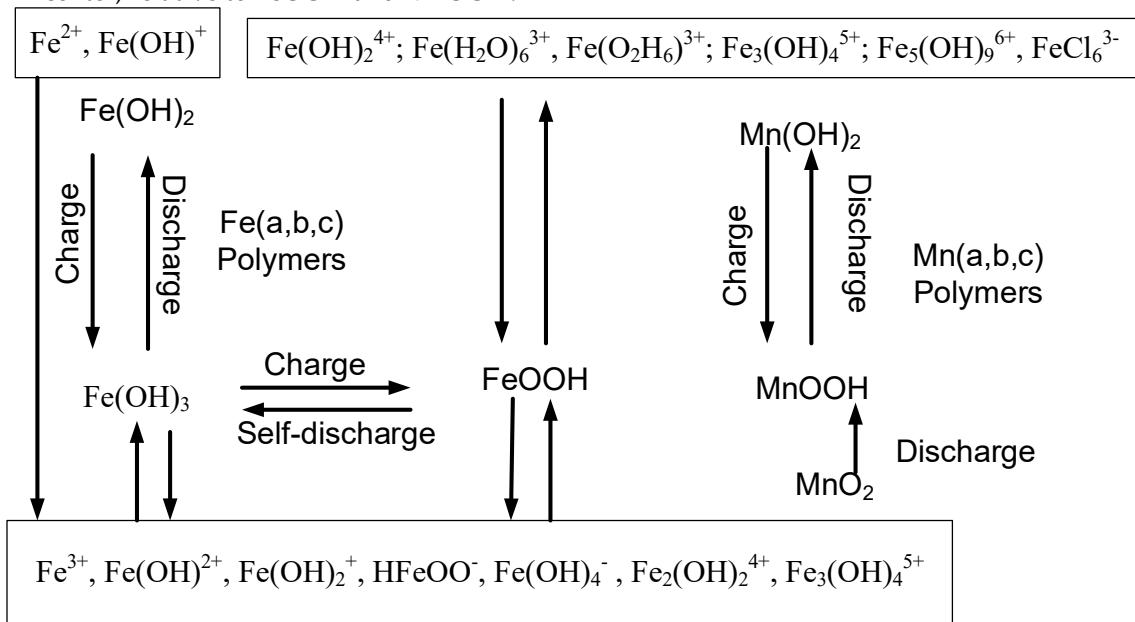


Figure S8. Electrochemical charge states associated with the metal polymers. The discharged oxidation number = 2 for Fe, Mn, and Zn. The charged oxidation number is 2 for Ca, Mg, 3 for Fe, Zn, Al, and 4 for Mn. Anodic reactions are associated with charging. Cathodic reactions are associated with discharging. MOOH and  $\text{MO}_x$  species (where  $x < 1$ ) can be visualised as capacitors.  $\text{M}^{n+}$  and  $\text{MO}$  species are discharged species.

### SA11 Implications for Supra-hydrophobic Sol-Gel Desalination Polymers

The typical sol-gel polymer (Figure S1) is positively charged, with substitutable OH<sup>-</sup> ions. If the Cl<sup>-</sup> ions are present as Cl<sup>-</sup>, or ClO<sup>-</sup>, ions then, they can substitute for the OH<sup>-</sup> ions (Figure S9). If the ions are present (in more acidic water) as HCl or HClO, then they will replace the OH<sup>-</sup> ions to produce an adsorbed species + water (Figure A8). The orientation of the polymers within the spheres is radial..

FeOOH and  $\text{Fe}(\text{O}_x\text{H}_y)^{n+/r}$  polymer crystals form initially, as isolate (typically hexagonal) plates, or spicules (needles) [152,153,154]. Aggregation, by crystal twinning and capture, initially creates a floral splay (a three dimensional V, or cone, shaped splay from an initial nucleus) or a dumbbell (where conical growth is in two directions) [153]. Aggregation initially creates a hydrogel, which allows for the formation of, spherical crystal growth, radiating from a defined nucleus [152]. FeOOH crystals formed in the presence of SO<sub>4</sub><sup>2-</sup> ions are dominantly spicular, or needle like [154]. In the presence of both SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup> ions, their dominant form is spherical [154]. This change has been interpreted, as reflecting a reduction in the surface charge of the crystallites, due to the presence of Cl<sup>-</sup> ions [154].

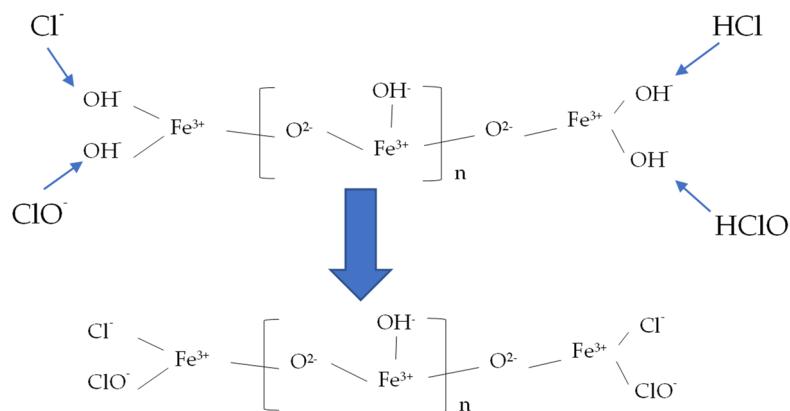
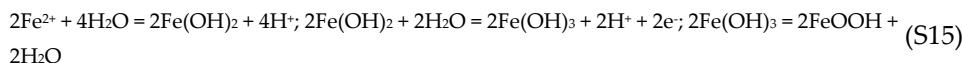


Figure S9. Schematic showing Cl<sup>-</sup> ion adsorption on edge located (Schottky) sites within the polymer

## SA12 Hydrogen Gas Formation

The basic polymer crystal structure, allows the polymer plates, to create a tethered slot for a Cl<sup>-</sup> ion, with a spectator Na<sup>+</sup> ion in its hydration shell (Figure S9). Reversing the polymer charge creates a tethered slot for a Na<sup>+</sup> ion, with a spectator Cl<sup>-</sup> ion in its hydration shell. The simple linear structure, allows a general migration of ions, to occur from the water body, to the center of the sphere and vice versa. The initial sphere centers contain water and H<sub>2</sub> gas.

The initial expulsion of water from the crystals, to the sphere center, is consistent with its super-hydrophobicity. The hydrogen gas, and water, result from the catalyzed water decomposition, as the FeOOH is formed. The formation of FeOOH from Fe<sup>2+</sup> requires the formation of both an Fe(OH)<sub>2</sub> intermediary and a Fe(OH)<sub>3</sub> intermediary (Equation S15). The byproducts of this process are 2H<sup>+</sup> ions, 1 e<sup>-</sup> (electron), and 1 unit of water. Expulsion of the H<sup>+</sup> ions, electrons and water from the super-hydrophobic radial spicules, into the fluid core, will result in the sphere fluid, containing hydrogen gas and acidic water (Equation S16). The expected initial molar ratio of the fluid core is 1 H<sub>2</sub> : 2H<sub>3</sub>O<sup>+</sup>. This implies that 3 moles of fluid within the spheres, will initially weigh 33 g, and occupy a volume of 22.446 L; i.e., the spheres, when initially formed, will be buoyant, and will float within the water body, as entrained particles.



The growth and aggregation of these polymer spheres (0.5 to 2 microns), results in a net loss of buoyancy with time, as the hydrogen is either expelled, or is adsorbed. The aggregated polymer spheres can form larger composite polymer spheres (60 – 1000 microns). These larger composite spheres can contain significant volumes of hydrogen gas (Figure S10a,b).

The polymer spheres (e.g., Fe<sup>0</sup>:Fe(b)@Al<sup>0</sup>), when they initially form, can fully encase the hydrogen gas bubble (Figure S10c). More commonly, the larger polymer spheres, will initially only partially cover the gas bubble with polymer (Figure S10c). The polymer spheres, rise, collide, and grow to form a solid mass constructed from a series of aggregated gas filled spheres (Figure S10d). In this example, the initial anodic reaction, at the sphere – waterbody interface is: 2Fe<sup>0</sup> = 2Fe<sup>2+</sup> + 4e<sup>-</sup>, and Al<sup>0</sup> + 3H<sub>2</sub>O = Al(OH)<sub>2</sub> + 3H<sup>+</sup> + 3e<sup>-</sup>. The cathodic reaction at the FeOOH – sphere core interface is: 13H<sup>+</sup> + 9e<sup>-</sup> + 2H<sub>2</sub>O = 4.5H<sub>2</sub>gas + 2H<sub>3</sub>O<sup>+</sup>.

Replacement of Fe<sup>0</sup> with Fe<sup>2+</sup> (this study), reduces the amount (moles) of H<sub>2</sub> produced per unit (mole) of Fe, by between 60% and 85%. This has three effects:

- (i) The produced gas filled spheres tend to be small, and fully encased by polymer;
- (ii) Growth is by collision, ion adsorption from the water body and Ostwald ripening;
- (iii) Aggregation occurs by collision of buoyant spheres (e.g., Figure S10d), and by collision of spheres, which have lost buoyancy (due to polymer growth or gas loss) and are settling.

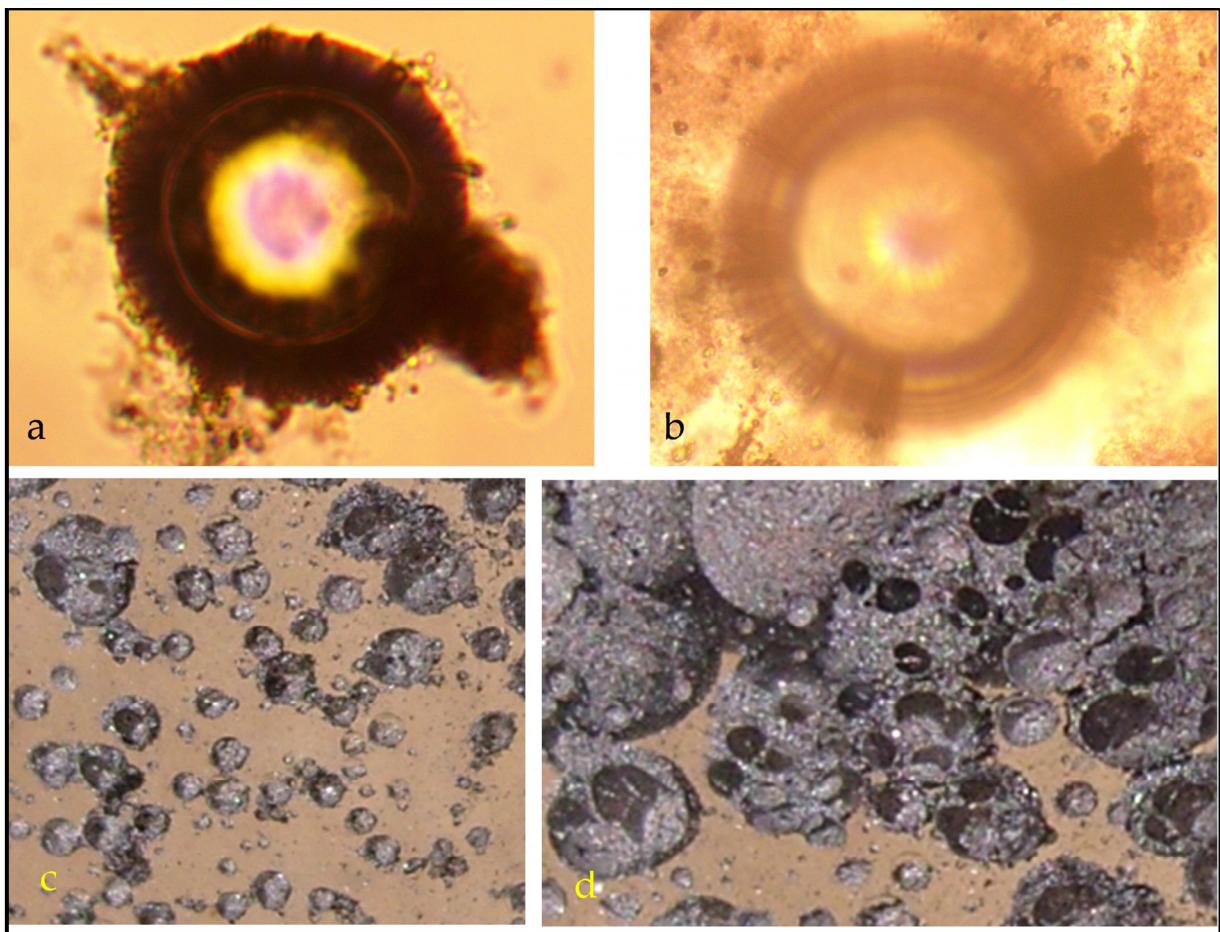


Figure S10. Examples of hydrogen bubbles associated with polymer formation. Water = seawater ( $\text{Cl}^- = 18.98 \text{ g L}^{-1}$ ;  $\text{Na}^+ = 15.5 \text{ g L}^{-1}$ ); Temperature = 288 K; Polymer constructed from:  $\text{FeSO}_4$  (4 g  $\text{L}^{-1}$ ) + gallic acid. Salinity after 5 h;  $\text{Cl}^- = 14.12 \text{ g L}^{-1}$ ;  $\text{Na}^+ = 11.6 \text{ g L}^{-1}$  (a), Hollow polymer sphere (62 microns diameter) composed of aggregated spherical units (1 – 2 microns in diameter) arranged radially around a fluid core (27.6 microns diameter) dominated by hydrogen. Field of view = 0.1 mm; (b), Hollow polymer sphere (110 micron diameter) composed of aggregated spherical units (1 – 2 microns in diameter) arranged radially around a fluid core (65.5 microns) dominated by hydrogen. Field of view = 0.167 mm; (c), Hydrogen filled  $\text{Fe}^0:\text{Fe}(b)@\text{Al}^0$  polymer spheres floating in saline water. Field of view = 10 mm; (d), Aggregating, floating, hydrogen filled  $\text{Fe}^0:\text{Fe}(b)@\text{Al}^0$  polymer spheres in saline water. Field of view = 10 mm; .

### SA13 Operation of Oxyhydroxide Metal Polymers

The prior studies have indicated that sol-gel precipitates, can be functionalized, and used to partially desalinate water. They operate by one or more of:

1. Approach 1: Direct reaction of the  $\text{Na}^+$  to form a ferrate and  $\text{Cl}^-$  to form an LDH; This limits the weight of the ion removed to between 10 and 30% of the weight of  $\text{Fe}^{n+}$  placed in the reaction environment;
2. Approach 2: Adsorption of one or more of  $\text{Na}^+$  and  $\text{Cl}^-$  ions, on to flocculate surfaces. This limits the weight of the ion removed to between 10 and 30% of the weight of  $\text{Fe}^{n+}$  placed in the reaction environment;
3. Approach 3: Adsorption of one or more of  $\text{Na}^+$  and  $\text{Cl}^-$  ions, onto flocculate surfaces, with subsequent migration through the walls of polymer spheres to the dead-end porosity, represented by the fluid core of the sphere. This process, uses Schottky and Frenkel defects to facilitate the ion transport. It leaves the fluid core volume unchanged as a function of time. It allows the weight of ions removed to exceed the weight of  $\text{Fe}^{n+}$  placed in the reaction environment;

4. Approach 4: Hydrated, poly-nuclear, metal hydroxyl ions ( $M(b)$ ) coalesce as hydrated polymer crystallites radiating from a sphere center. Direct interaction with  $Na^+$  ions,  $Cl^-$  ions, and other ions, contained in the water body, surrounding the polymer sphere, results in their incorporation, within the hydrated polymer crystallites. Ostwald ripening at the sphere center, results in dissolution of the hydrated polymer crystallites, to produce an expanding fluid core. Polymer dissolution forms mono-nuclear metal hydroxyl ions, or metal ions, plus  $Na^+$  ions and  $Cl^-$  ions, plus other incorporate ions within the polymer, which have been released, plus water. This process results in, a gradual increase, in the sphere core diameter size, coupled with a gradual increase in sphere diameter. The hydrated, mono-nuclear, metal hydroxyl ions, or metal ions ( $M(a)$ ), migrate into gaps in the expanding sphere and may migrate to its outer margin. The enlarging sphere core, becomes gradually filled, with released ions and water over time. This process results in the fluid core volume increasing as a function of time. It allows the weight of ions removed to exceed the weight of  $Fe^{n+}$  placed in the reaction environment;

Approach 3 assumes that the polymers are hydrophobic, and allow passage of  $Na^+$  and  $Cl^-$  ions through the polymer, but not  $H_2O$ . Approach 4 either (i), assumes that the polymers are hydrophilic, and allow  $H_2O$  to flow from the water body to the sphere center, but does not allow ions (including  $Na^+$  and  $Cl^-$ ) concentrated in the sphere center, to migrate through the polymer to the water body, or (ii), assumes that the polymers are hydrophobic, and do not allow  $H_2O$  to flow from the water body, to the sphere center, but do allow ions (including  $Na^+$  and  $Cl^-$ ) located in the water body, to migrate through the polymer, to the sphere center.

The 87 different sol-gel metal polymer combinations (Figure S11), indicated that there is no simple, universal, relationship between  $Na^+$  removal, or  $Cl^-$  removal (selectivity). However, as demonstrated in this study, using  $n\text{-Fe}(a,b,c)@\text{MnO}_2$  polymers, it may be possible to develop polymer specific relationships between redox conditions and  $Na^+$  removal, or  $Cl^-$  removal. These relationships, when present (or definable), may also be specific to a target saline water body.

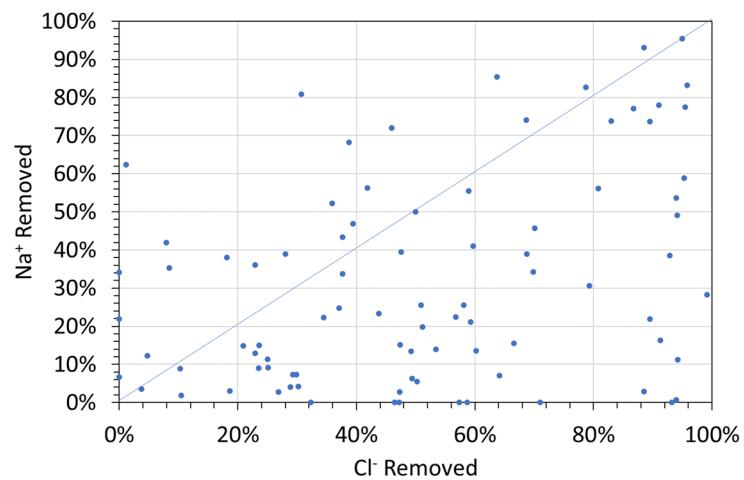


Figure S11. Ion removal selectivity associated with Sol-Gel Polymer Desalination.

## Supplementary Information Section SB: Data Used In This Study

Table S1. Feed water and product water ion concentrations and ion molar ratios. When the molar ratio = 1.0, all the Na<sup>+</sup> and Cl<sup>-</sup> ions could form NaCl. When the molar ratio is <1, then some of the Na<sup>+</sup> ions are present as NaOH or as another soluble Na mineral (e.g., NaHCO<sub>3</sub>). When the molar ratio is >1, then some of the Cl<sup>-</sup> ions are present as HClO, ClO<sup>-</sup>, HCl or as another soluble Cl<sup>-</sup> mineral (e.g., CaCl<sub>2</sub>).

Trial	Feed			Product						
	Cl <sup>-</sup> , g/L	Na <sup>+</sup> , g/L	Total, g/L	Moles Cl:Na	NaCl, g/L	Cl <sup>-</sup> , g/L	Na <sup>+</sup> , g/L	Total, g/L		
F1	26.55	17.21	43.76	0.96	42.63	1.35	0.78	2.13	1.07	2.08
F2	26.55	17.21	43.76	0.96	42.63	5.66	2.97	8.63	1.18	8.17
F3	26.55	17.21	43.76	0.96	42.63	1.55	8.75	10.30	0.11	8.92
F4	26.55	17.21	43.76	0.96	42.63	9.64	2.50	12.14	2.39	10.68
F5	22.11	14.43	36.54	0.95	35.45	1.51	14.43	15.94	0.06	14.53
F6	22.11	14.43	36.54	0.95	35.45	1.93	12.07	14.00	0.10	12.26
F7	22.11	14.43	36.54	0.95	35.45	1.04	5.93	6.97	0.11	6.04
F8	22.11	14.43	36.54	0.95	35.45	11.67	14.43	26.10	0.50	20.29
F9	22.11	14.43	36.54	0.95	35.45	6.61	7.84	14.45	0.52	11.30
F10	22.11	14.43	36.54	0.95	35.45	1.00	3.25	4.25	0.19	3.44
F11	22.11	14.43	36.54	0.95	35.45	1.33	14.33	15.66	0.06	14.41
F12	22.11	14.43	36.54	0.95	35.45	1.29	12.82	14.11	0.06	12.90
F13	22.11	14.43	36.54	0.95	35.45	1.99	3.18	5.17	0.39	3.95
F14	22.11	14.43	36.54	0.95	35.45	3.76	3.77	7.53	0.62	6.10
F15	23.22	19.83	43.05	0.73	36.70	0.97	3.32	4.29	0.18	3.50
F16	19.52	13.36	32.88	0.91	31.06	6.12	3.46	9.58	1.10	9.27
F17	26.40	15.83	42.23	1.03	41.70	3.03	1.09	4.12	1.73	3.66
F18	26.11	19.57	45.68	0.83	41.19	18.10	3.75	21.85	3.00	19.35
F19	101.48	10.51	111.99	5.99	103.23	7.27	6.46	13.73	0.70	11.54
F20	47.76	21.49	69.25	1.38	63.34	9.15	9.42	18.57	0.60	14.94
F21	25.47	23.95	49.42	0.66	40.76	25.18	9.00	34.18	1.74	30.36
F22	17.40	18.73	36.13	0.58	28.76	8.71	9.37	18.08	0.58	14.39
F23	19.94	8.47	28.41	1.46	25.74	6.01	5.57	11.58	0.67	9.59
F24	10.44	6.33	16.77	1.02	16.62	4.51	4.91	9.42	0.57	7.48
F25	2.76	1.23	3.99	1.39	3.64	0.80	1.23	2.03	0.40	1.55
F26	3.49	1.85	5.34	1.17	5.07	1.39	1.60	2.99	0.54	2.35
F27	4.40	1.79	6.19	1.53	5.57	2.32	1.74	4.06	0.83	3.66
F28	9.40	4.98	14.38	1.17	13.65	5.86	3.30	9.16	1.10	8.85
F29	12.14	3.13	15.27	2.41	13.44	6.50	3.13	9.63	1.29	8.93
F30	22.11	14.33	36.44	0.96	35.50	1.34	6.65	7.99	0.13	6.82
F31	22.11	14.33	36.44	0.96	35.50	2.32	3.76	6.08	0.38	4.65
F32	22.11	14.33	36.44	0.96	35.50	2.93	3.29	6.22	0.55	4.91
F33	30.14	18.38	48.52	1.02	48.20	12.46	18.38	30.84	0.42	23.62
F34	30.14	18.38	48.52	1.02	48.20	24.67	11.38	36.05	1.35	33.13
F35	30.14	18.38	48.52	1.02	48.20	9.43	11.22	20.65	0.52	16.14
F36	11.92	5.41	17.33	1.37	15.88	4.86	4.27	9.13	0.71	7.70
F37	3.07	1.41	4.48	1.35	4.11	1.50	1.13	2.63	0.82	2.37
F38	8.14	4.31	12.45	1.17	11.82	5.22	2.06	7.28	1.57	6.53
F39	30.47	10.30	40.77	1.84	36.08	30.47	6.78	37.25	2.79	32.90
F40	30.47	10.30	40.77	1.84	36.08	23.49	6.58	30.07	2.22	26.46
F41	25.36	16.44	41.80	0.96	40.72	13.35	13.95	27.30	0.59	21.88
F42	14.50	9.99	24.49	0.90	23.05	7.36	8.65	16.01	0.53	12.54
F43	14.50	9.99	24.49	0.90	23.05	7.34	9.36	16.70	0.49	12.93
F44	1.22	1.52	2.74	0.50	2.13	0.64	0.92	1.56	0.43	1.20
F45	10.57	10.10	20.67	0.65	16.96	2.19	7.01	9.20	0.19	7.43
F46	57.04	45.57	102.61	0.78	89.88	33.17	19.96	53.13	1.03	52.52
F47	2.19	7.01	9.20	0.19	7.43	0.23	5.48	5.71	0.03	5.49
F48	33.17	19.96	53.13	1.03	52.52	13.38	11.78	25.16	0.70	21.21
F49	11.54	16.77	28.31	0.43	21.70	10.36	15.29	25.65	0.42	19.65
F50	11.54	16.77	28.31	0.43	21.70	9.13	14.28	23.41	0.40	17.90
F51	24.72	19.50	44.22	0.79	38.95	0.21	13.99	14.20	0.01	13.99
F52	1.48	0.70	2.18	1.31	2.01	0.17	0.68	0.85	0.16	0.71
F53	2.06	1.53	3.59	0.84	3.25	1.35	1.19	2.54	0.70	2.14
F54	19.50	14.75	34.25	0.82	30.75	10.97	11.31	22.28	0.60	17.91
F55	19.50	14.75	34.25	0.82	30.75	12.16	8.35	20.51	0.90	19.34
F56	57.26	44.14	101.40	0.81	90.24	26.66	37.97	64.63	0.44	49.59

F57	57.26	44.14	101.40	0.81	90.24	41.19	26.95	68.14	0.95	66.02
F58	19.17	9.76	28.93	1.22	27.18	11.61	5.19	16.80	1.39	15.35
F59	19.17	9.76	28.93	1.22	27.18	7.87	4.34	12.21	1.13	11.73
F60	1.24	0.82	2.06	0.94	1.98	0.76	0.26	1.02	1.81	0.90
F61	1.24	0.82	2.06	0.94	1.98	0.67	0.23	0.90	1.81	0.80
F62	24.67	13.23	37.90	1.16	36.10	18.86	11.25	30.11	1.04	29.67
F63	24.67	13.23	37.90	1.16	36.10	22.60	8.56	31.16	1.64	27.82
F64	19.30	11.18	30.48	1.07	29.74	14.88	9.73	24.61	0.95	23.85
F65	19.30	11.18	30.48	1.07	29.74	9.60	10.56	20.16	0.56	15.98
F66	49.31	15.40	64.71	1.99	57.06	44.17	15.11	59.28	1.81	52.50
F67	49.31	15.40	64.71	1.99	57.06	47.47	14.85	62.32	1.98	54.96
F68	21.87	17.74	39.61	0.77	34.47	9.33	17.74	27.07	0.33	20.79
F69	21.87	17.74	39.61	0.77	34.47	7.32	14.99	22.31	0.30	17.21
F70	14.50	16.04	30.54	0.56	24.17	10.88	14.22	25.10	0.47	19.39
F71	14.50	16.04	30.54	0.56	24.17	7.12	11.95	19.07	0.37	14.58
F72	28.10	14.73	42.83	1.18	40.54	22.86	14.28	37.14	0.99	36.99
F73	28.10	14.73	42.83	1.18	40.54	19.61	14.12	33.73	0.86	31.02
F74	10.92	7.57	18.49	0.90	17.35	10.92	5.91	16.83	1.15	16.07
F75	10.92	7.57	18.49	0.90	17.35	4.58	5.64	10.22	0.50	7.95
F76	6.41	3.96	10.37	1.00	10.35	4.69	3.85	8.54	0.76	7.40
F77	6.41	3.96	10.37	1.00	10.35	2.30	3.68	5.98	0.39	4.57
F78	10.59	9.47	20.06	0.69	16.82	10.59	8.84	19.43	0.74	16.71
F79	10.59	9.47	20.06	0.69	16.82	6.66	7.12	13.78	0.58	10.99
F80	11.48	15.61	27.09	0.46	20.85	10.94	13.70	24.64	0.50	19.12
F81	11.48	15.61	27.09	0.46	20.85	10.57	9.06	19.63	0.72	16.71
F82	9.60	7.54	17.14	0.79	15.13	7.34	6.86	14.20	0.66	11.73
F83	9.60	7.54	17.14	0.79	15.13	7.19	6.85	14.04	0.65	11.53
F84	9.68	9.50	19.18	0.63	15.62	6.79	8.81	15.60	0.48	12.06
F85	9.68	9.50	19.18	0.63	15.62	6.85	8.80	15.65	0.48	12.11
F86	7.96	7.74	15.70	0.64	12.82	5.66	7.43	13.09	0.47	10.11
F87	7.96	7.74	15.70	0.64	12.82	5.39	7.74	13.13	0.43	10.07

Table S2. Feed water and product water redox parameters. hrs = reaction time between measurement.

Trial	Feed			Product					Change in PSE
	pH	Eh, V	PSE, V	hrs	pH	Eh, V	PSE, V	PSE <sub>n</sub> , V	
F1	8.31	0.37	0.0448	241.00	11.85	-0.05	-0.00380	0.01374	-0.01753
F2	8.31	0.37	0.0448	241.00	12.23	-0.07	-0.00605	-0.02499	0.01894
F3	8.31	0.37	0.0448	241.00	9.31	0.19	0.02073	0.01438	0.00635
F4	8.31	0.37	0.0448	241.00	12.54	-0.10	-0.00781	-0.02775	0.01994
F5	8.69	0.04	0.0045	0.10	2.90	0.87	0.29931	0.41731	-0.11800
F6	8.69	0.04	0.0045	0.10	6.20	0.02	0.00387	0.02761	-0.02374
F7	8.69	0.04	0.0045	216.00	9.37	0.17	0.01836	0.01407	0.00429
F8	8.69	0.04	0.0045	0.10	3.42	0.81	0.23538	0.32645	-0.09107
F9	8.69	0.04	0.0045	0.10	6.91	0.03	0.00420	0.01942	-0.01522
F10	8.69	0.04	0.0045	216.00	10.11	0.06	0.00603	-0.00227	0.00830
F11	8.69	0.04	0.0045	0.10	3.83	0.72	0.18877	0.26377	-0.07499
F12	8.69	0.04	0.0045	0.10	6.57	0.12	0.01781	0.03688	-0.01907
F13	8.69	0.04	0.0045	216.00	10.34	0.04	0.00358	-0.00585	0.00943
F14	8.69	0.04	0.0045	96.00	11.18	0.03	0.00233	-0.01084	0.01316
F15	8.57	0.25	0.0291	24.00	12.28	-0.10	-0.00839	-0.02624	0.01786
F16	8.75	0.25	0.0281	24.00	12.16	-0.10	-0.00806	-0.02463	0.01657
F17	8.70	0.23	0.0268	24.00	12.15	-0.35	-0.02840	-0.04518	0.01678
F18	9.33	0.18	0.0190	48.00	11.96	-0.03	-0.00217	-0.01517	0.01300
F19	9.49	0.15	0.0162	24.00	9.91	0.28	0.02866	0.02615	0.00250
F20	9.37	0.27	0.0283	24.00	12.33	-0.05	-0.00365	-0.01784	0.01419
F21	9.17	0.23	0.0250	24.00	11.72	-0.04	-0.00299	-0.01585	0.01286
F22	9.58	0.17	0.0181	24.00	11.22	0.06	0.00561	-0.00302	0.00864
F23	9.54	0.23	0.0239	24.00	13.10	-0.08	-0.00618	-0.02224	0.01606
F24	9.51	0.18	0.0190	24.00	11.87	0.00	-0.00034	-0.01209	0.01175
F25	9.78	0.15	0.0155	24.00	13.08	-0.08	-0.00612	-0.02103	0.01491
F26	9.76	0.14	0.0148	24.00	12.42	-0.04	-0.00298	-0.01564	0.01266

F27	9.54	0.16	0.0170	24.00	12.51	-0.02	-0.00184	-0.01587	0.01403
F28	9.06	0.20	0.0216	24.00	12.82	-0.08	-0.00601	-0.02334	0.01733
F29	9.05	0.22	0.0238	24.00	12.87	-0.07	-0.00513	-0.02267	0.01754
F30	8.69	0.26	0.0293	1344.00	7.97	0.35	0.04442	0.04976	-0.00534
F31	8.69	0.26	0.0293	1344.00	8.66	0.28	0.03256	0.03277	-0.00020
F32	8.69	0.26	0.0293	1398.00	9.62	0.18	0.01913	0.01341	0.00571
F33	8.81	0.34	0.0390	0.08	6.19	0.33	0.05267	0.07768	-0.02501
F34	8.81	0.34	0.0390	0.08	6.61	0.17	0.02572	0.04539	-0.01967
F35	8.81	0.34	0.0390	72.00	12.96	-0.13	-0.01011	-0.02903	0.01892
F36	10.35	0.08	0.0081	48.00	13.07	-0.14	-0.01064	-0.02293	0.01230
F37	10.42	0.07	0.0071	48.00	12.36	-0.10	-0.00841	-0.01769	0.00928
F38	9.07	0.16	0.0175	48.00	7.17	0.15	0.02134	0.03700	-0.01566
F39	8.56	0.29	0.0338	24.00	7.34	0.01	0.00177	0.01159	-0.00982
F40	8.56	0.29	0.0338	24.00	7.35	0.09	0.01197	0.02170	-0.00973
F41	8.55	0.26	0.0303	96.00	11.98	-0.07	-0.00593	-0.02285	0.01692
F42	10.31	0.08	0.0079	96.00	12.75	-0.11	-0.00875	-0.02006	0.01131
F43	10.17	0.09	0.0084	24.00	12.78	-0.11	-0.00850	-0.02057	0.01207
F44	9.13	0.16	0.0170	24.00	10.49	0.13	0.01209	0.00443	0.00766
F45	9.86	0.19	0.0190	72.00	11.35	0.04	0.00320	-0.00456	0.00776
F46	8.64	0.30	0.0351	72.00	12.06	-0.30	-0.02498	-0.04174	0.01676
F47	11.35	0.04	0.0032	48.00	6.77	0.16	0.02335	0.06334	-0.03998
F48	12.06	-0.30	-0.0250	24.00	6.51	0.18	0.02691	0.07730	-0.05038
F49	12.68	-0.47	-0.0374	24.00	9.23	0.24	0.02563	0.04772	-0.02209
F50	12.68	-0.47	-0.0374	24.00	10.58	0.11	0.01020	0.02193	-0.01173
F51	9.78	0.17	0.0176	72.00	12.41	-0.07	-0.00575	-0.01827	0.01252
F52	10.67	0.07	0.0067	24.00	11.47	0.04	0.00310	-0.00103	0.00412
F53	9.91	0.17	0.0168	24.00	10.31	0.19	0.01820	0.01590	0.00229
F54	9.96	0.20	0.0202	24.00	9.58	0.21	0.02209	0.02443	-0.00234
F55	9.96	0.20	0.0202	24.00	10.13	0.16	0.01565	0.01465	0.00099
F56	9.53	0.23	0.0237	24.00	6.11	0.59	0.09712	0.13020	-0.03308
F57	9.53	0.23	0.0237	24.00	12.55	-0.03	-0.00243	-0.01665	0.01422
F58	8.86	0.27	0.0309	24.00	5.51	0.66	0.11902	0.15495	-0.03593
F59	8.86	0.27	0.0309	24.00	6.55	0.56	0.08482	0.10567	-0.02084
F60	8.29	0.33	0.0393	24.00	8.12	0.37	0.04538	0.04662	-0.00124
F61	8.29	0.33	0.0393	24.00	9.10	0.29	0.03232	0.02706	0.00526
F62	8.49	0.36	0.0424	48.00	7.46	0.47	0.06298	0.07114	-0.00816
F63	8.49	0.36	0.0424	48.00	7.56	0.46	0.06101	0.06828	-0.00727
F64	8.13	0.41	0.0498	72.00	6.68	0.53	0.07919	0.09202	-0.01283
F65	8.13	0.41	0.0498	72.00	12.86	-0.06	-0.00481	-0.02654	0.02174
F66	9.80	0.19	0.0198	24.00	8.92	0.33	0.03698	0.04281	-0.00583
F67	9.80	0.19	0.0198	24.00	8.89	0.33	0.03767	0.04372	-0.00605
F68	8.84	0.32	0.0359	24.00	5.42	0.69	0.12672	0.16401	-0.03729
F69	8.84	0.32	0.0359	24.00	11.12	0.19	0.01677	0.00465	0.01212
F70	8.72	0.32	0.0368	24.00	5.64	0.63	0.11087	0.14314	-0.03227
F71	8.72	0.32	0.0368	24.00	12.68	-0.01	-0.00063	-0.01909	0.01846
F72	8.68	0.31	0.0355	24.00	6.34	0.46	0.07229	0.09410	-0.02181
F73	8.68	0.31	0.0355	24.00	12.92	-0.04	-0.00323	-0.02262	0.01940
F74	9.60	0.23	0.0240	96.00	7.12	-0.07	-0.00954	0.01105	-0.02059
F75	9.60	0.23	0.0240	96.00	12.69	-0.64	-0.05009	-0.06448	0.01439
F76	8.96	0.30	0.0331	24.00	7.83	0.42	0.05307	0.06159	-0.00853
F77	8.96	0.30	0.0331	24.00	12.27	0.06	0.00472	-0.01122	0.01594
F78	8.92	0.34	0.0385	24.00	7.43	0.08	0.01020	0.02205	-0.01185
F79	8.92	0.34	0.0385	24.00	7.45	0.16	0.02181	0.03347	-0.01166
F80	9.06	0.42	0.0467	24.00	7.41	0.08	0.01135	0.02451	-0.01316
F81	9.06	0.42	0.0467	24.00	9.69	0.00	-0.00037	-0.00421	0.00384
F82	9.79	0.36	0.0368	24.00	9.49	0.38	0.03988	0.04175	-0.00187
F83	9.79	0.36	0.0368	24.00	10.37	0.30	0.02871	0.02540	0.00331
F84	9.67	0.36	0.0373	24.00	9.54	0.36	0.03818	0.03898	-0.00081
F85	9.67	0.36	0.0373	24.00	10.26	0.28	0.02738	0.02398	0.00340
F86	9.80	0.35	0.0354	24.00	9.13	0.39	0.04218	0.04652	-0.00434
F87	9.80	0.35	0.0354	24.00	10.53	0.24	0.02272	0.01862	0.00410

Table S3. Polymer Ingredients ( $\text{Fe}^{2+}$  +  $\text{Ca}^{2+}$  +  $\text{Mg}^{2+}$  +  $\text{Mn}^{4+}$ ). Units are g L<sup>-1</sup>. Grey highlight = formulations which dominantly removed Cl<sup>-</sup> ions; Gold highlight = formulations which dominantly removed Na<sup>+</sup> ions.

Trial	Change in pH	Cl <sup>-</sup> Removal	Na <sup>+</sup> Removal	PSE Change	Category	FeSO <sub>4</sub>	MgSO <sub>4</sub>	MgCO <sub>3</sub>	CaO	CaCO <sub>3</sub>	MnO <sub>2</sub>
F1	3.54	94.9%	95.5%	-0.02	A7d	1			1.67		2.52
F2	3.92	78.7%	82.7%	0.02	A7f	1			1.67		2.52
F3	1.00	94.2%	49.2%	0.01	A7e	1		2.96	1.67		2.52
F4	4.23	63.7%	85.5%	0.02	A7d	1			3.34		2.52
F5	-5.79	93.2%	0.0%	-0.12	A3	1					
F6	-2.49	91.3%	16.4%	-0.02	A3a	1			1.67	1.36	
F7	0.68	95.3%	58.9%	0.00	A3b	1			3.34	1.36	2.52
F8	-5.27	47.2%	0.0%	-0.09	A4	1					
F9	-1.78	70.1%	45.7%	-0.02	A4b	1			1.67	1.36	
F10	1.42	95.5%	77.5%	0.01	A4c	1			1.67	2.71	2.52
F11	-4.86	94.0%	0.7%	-0.07	A5	1					
F12	-2.12	94.2%	11.2%	-0.02	A5b	1			1.67	1.36	
F13	1.65	91.0%	78.0%	0.01	A5c	1			1.67	4.07	2.52
F14	2.49	83.0%	73.9%	0.01	A3c	1			1.67	1.36	2.52
F15	3.71	95.8%	83.3%	0.02	A3d	2			3.34		
F16	3.41	68.6%	74.1%	0.02	A4a	2			3.34		
F17	3.45	88.5%	93.1%	0.02	A5a	2			3.34		
F18	2.63	30.7%	80.8%	0.01	A3d	2			3.34		
F19	0.42	92.8%	38.5%	0.00	A5a	2			3.34		
F20	2.96	80.8%	56.2%	0.01	A6f	2			3.34	2.71	
F21	2.55	1.1%	62.4%	0.01	A7g	2		1.48	3.34	2.71	2.52
F22	1.64	49.9%	50.0%	0.01	A6g	2		1.48	3.34	2.71	2.52
F23	3.56	69.9%	34.2%	0.02	A5a	2			3.34		
F24	2.36	56.8%	22.4%	0.01	A6i	2			3.34		
F25	3.30	71.0%	0.0%	0.01	A4a	2			3.34		
F26	2.66	60.2%	13.5%	0.01	A6h	2			3.34		
F27	2.97	47.3%	2.8%	0.01	A6e	2			3.34		
F28	3.76	37.7%	33.7%	0.02	A7	2			3.34		
F29	3.82	46.5%	0.0%	0.02	A7	1			1.67		
F30	-0.72	93.9%	53.6%	-0.01	A7c	0.69		1.48	3.34		2.52
F31	-0.03	89.5%	73.8%	0.00	A7b	0.8			3.34		2.52
F32	0.93	86.7%	77.0%	0.01	A7a	0.8			3.34	2.71	
F33	-2.62	58.7%	0.0%	-0.03	A2a	1.42			3.34		
F34	-2.20	18.1%	38.1%	-0.02	A2a	1.42			1.67		
F35	4.15	68.7%	39.0%	0.02	A2a	1.42			3.34		
F36	2.72	59.2%	21.1%	0.01	B1				3.34		
F37	1.94	51.1%	19.9%	0.01	B1				3.34		
F38	-1.90	35.9%	52.2%	-0.02	A6	1.42					
F39	-1.22	0.0%	34.2%	-0.01	A6	1.42					
F40	-1.21	22.9%	36.1%	-0.01	A6b	1.42			3.34		
F41	3.43	47.4%	15.1%	0.02	B2a				1.67		2.52
F42	2.44	49.2%	13.4%	0.01	B2a				1.67		2.52
F43	2.61	49.4%	6.3%	0.01	B2				1.67		
F44	1.36	47.5%	39.5%	0.01	C4		1.51				
F45	1.49	79.3%	30.6%	0.01	A6d	1.42	1.51		1.67		
F46	3.42	41.8%	56.2%	0.02	A6c	1.42	0.76		1.67		
F47	-4.58	89.5%	21.8%	-0.04	A1	1.42					
F48	-5.55	59.7%	41.0%	-0.05	A2	1.42					
F49	-3.45	10.2%	8.8%	-0.02	A8	1.42	1.51				
F50	-2.10	20.9%	14.8%	-0.01	A2b	1.42	1.51				
F51	2.63	99.2%	28.3%	0.01	A8h	2.84	1.51		3.34	2.71	
F52	0.80	88.5%	2.9%	0.00	A8g	5.68	1.51		1.67		
F53	0.40	34.5%	22.2%	0.00	C1		1.51				
F54	-0.38	43.7%	23.3%	0.00	C1		3.02				
F55	0.17	37.6%	43.4%	0.00	C2		3.02		1.67		
F56	-3.42	53.4%	14.0%	-0.03	C3		1.51				
F57	3.02	28.1%	38.9%	0.01	C2a		1.51		1.67		
F58	-3.35	39.4%	46.8%	-0.04	C3		3.02				
F59	-2.31	58.9%	55.5%	-0.02	C2a		3.02		1.67		
F60	-0.17	38.7%	68.3%	0.00	A8i	1.42	3.02		1.67	1.36	2.52
F61	0.81	46.0%	72.0%	0.01	A8i	1.42	3.02		3.34	1.36	2.52
F62	-1.03	23.6%	15.0%	-0.01	D1a				1.67		
F63	-0.93	8.4%	35.3%	-0.01	B3						
F64	-1.45	22.9%	13.0%	-0.01	D1a						

F65	4.73	50.3%	5.5%	0.02	B3		3.34		
F66	-0.88	10.4%	1.9%	-0.01	B3a		1.67	1.36	
F67	-0.91	3.7%	3.6%	-0.01	D1				
F68	-3.42	57.3%	0.0%	-0.04	C3	1.51			
F69	2.28	66.5%	15.5%	0.01	C2a	1.51		1.67	
F70	-3.08	25.0%	11.3%	-0.03	C3	1.51			
F71	3.96	50.9%	25.5%	0.02	C2a	1.51		1.67	
F72	-2.34	18.6%	3.1%	-0.02	C3	1.51			
F73	4.24	30.2%	4.1%	0.02	C2a	1.51		1.67	
F74	-2.48	0.0%	21.9%	-0.02	A8c	1.42	1.51		
F75	3.09	58.1%	25.5%	0.01	A8f	1.42	1.51		1.67
F76	-1.13	26.8%	2.8%	-0.01	A8b	1.42	1.51		
F77	3.31	64.1%	7.1%	0.02	A8e	1.42	1.51		1.67
F78	-1.49	0.0%	6.7%	-0.01	A8c	1.42	1.51		1.36
F79	-1.47	37.1%	24.8%	-0.01	A8d	1.42	1.51		
F80	-1.65	4.7%	12.2%	-0.01	A8c	1.42	1.51		
F81	0.63	7.9%	42.0%	0.00	A8d	1.42	1.51		
F82	-0.30	23.5%	9.0%	0.00	C1		2.27		
F83	0.58	25.1%	9.2%	0.00	C4a		2.27		
F84	-0.13	29.9%	7.3%	0.00	C5		1.51		2.52
F85	0.59	29.2%	7.4%	0.00	C4b		1.51		2.52
F86	-0.67	28.9%	4.0%	0.00	D1b				2.52
F87	0.73	32.3%	0.0%	0.00	A8a	1.42			1.67

Table S12. Polymer Ingredients ( $K^+ + Zn^{2+} + Al^{3+}$ ). Units are g L<sup>-1</sup>. Grey highlight = formulations which dominantly removed Cl<sup>-</sup> ions; Gold highlight = formulations which dominantly removed Na<sup>+</sup> ions.

Trial	Change in pH	Cl <sup>-</sup> Removal	Na <sup>+</sup> Removal	PSE Change	K <sub>2</sub> SO <sub>4</sub>	K-Feldspar	ZnO	Al <sup>0</sup>	Al <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	Al(OH) <sub>3</sub>
F1	3.54	94.9%	95.5%	-0.02							
F2	3.92	78.7%	82.7%	0.02	1.22						
F3	1.00	94.2%	49.2%	0.01							
F4	4.23	63.7%	85.5%	0.02							
F5	-5.79	93.2%	0.0%	-0.12							
F6	-2.49	91.3%	16.4%	-0.02	1.22						
F7	0.68	95.3%	58.9%	0.00	1.22						
F8	-5.27	47.2%	0.0%	-0.09							
F9	-1.78	70.1%	45.7%	-0.02	1.22						
F10	1.42	95.5%	77.5%	0.01	1.22						
F11	-4.86	94.0%	0.7%	-0.07							
F12	-2.12	94.2%	11.2%	-0.02	1.22						
F13	1.65	91.0%	78.0%	0.01	1.22						
F14	2.49	83.0%	73.9%	0.01	1.22						
F15	3.71	95.8%	83.3%	0.02			11.22				
F16	3.41	68.6%	74.1%	0.02			11.22				
F17	3.45	88.5%	93.1%	0.02			11.22				
F18	2.63	30.7%	80.8%	0.01			5.61				
F19	0.42	92.8%	38.5%	0.00			5.61				
F20	2.96	80.8%	56.2%	0.01			5.61				
F21	2.55	1.1%	62.4%	0.01			2.81				
F22	1.64	49.9%	50.0%	0.01			5.61				
F23	3.56	69.9%	34.2%	0.02			5.61				
F24	2.36	56.8%	22.4%	0.01			5.61				
F25	3.30	71.0%	0.0%	0.01			5.61				
F26	2.66	60.2%	13.5%	0.01			5.61				
F27	2.97	47.3%	2.8%	0.01			5.61				
F28	3.76	37.7%	33.7%	0.02							
F29	3.82	46.5%	0.0%	0.02							
F30	-0.72	93.9%	53.6%	-0.01	0.61						
F31	-0.03	89.5%	73.8%	0.00							
F32	0.93	86.7%	77.0%	0.01							
F33	-2.62	58.7%	0.0%	-0.03							
F34	-2.20	18.1%	38.1%	-0.02							
F35	4.15	68.7%	39.0%	0.02							
F36	2.72	59.2%	21.1%	0.01							
F37	1.94	51.1%	19.9%	0.01							
F38	-1.90	35.9%	52.2%	-0.02			5.61				
F39	-1.22	0.0%	34.2%	-0.01			5.61				

F40	-1.21	22.9%	36.1%	-0.01		8.42	1.36	
F41	3.43	47.4%	15.1%	0.02	1.99			
F42	2.44	49.2%	13.4%	0.01	1.99			
F43	2.61	49.4%	6.3%	0.01	1.99			
F44	1.36	47.5%	39.5%	0.01	1.22	1.99		
F45	1.49	79.3%	30.6%	0.01	1.99	2.81		
F46	3.42	41.8%	56.2%	0.02	1.22	2.81		
F47	-4.58	89.5%	21.8%	-0.04				
F48	-5.55	59.7%	41.0%	-0.05				
F49	-3.45	10.2%	8.8%	-0.02				
F50	-2.10	20.9%	14.8%	-0.01	1.22			
F51	2.63	99.2%	28.3%	0.01			2.5	
F52	0.80	88.5%	2.9%	0.00				
F53	0.40	34.5%	22.2%	0.00				1.2
F54	-0.38	43.7%	23.3%	0.00				2.4
F55	0.17	37.6%	43.4%	0.00				3.6
F56	-3.42	53.4%	14.0%	-0.03				1.34
F57	3.02	28.1%	38.9%	0.01				1.2
F58	-3.35	39.4%	46.8%	-0.04				1.34
F59	-2.31	58.9%	55.5%	-0.02				1.34
F60	-0.17	38.7%	68.3%	0.00	2.81			3.6
F61	0.81	46.0%	72.0%	0.01	2.81			1.34
F62	-1.03	23.6%	15.0%	-0.01	2.81			1.34
F63	-0.93	8.4%	35.3%	-0.01	2.81			1.34
F64	-1.45	22.9%	13.0%	-0.01	4.22			1.8
F65	4.73	50.3%	5.5%	0.02	4.22			1.8
F66	-0.88	10.4%	1.9%	-0.01	2.81			1.34
F67	-0.91	3.7%	3.6%	-0.01	2.81		0	1.2
F68	-3.42	57.3%	0.0%	-0.04				1.34
F69	2.28	66.5%	15.5%	0.01				1.2
F70	-3.08	25.0%	11.3%	-0.03				1.34
F71	3.96	50.9%	25.5%	0.02				1.2
F72	-2.34	18.6%	3.1%	-0.02				1.34
F73	4.24	30.2%	4.1%	0.02				1.34
F74	-2.48	0.0%	21.9%	-0.02			0	1.2
F75	3.09	58.1%	25.5%	0.01			0	1.2
F76	-1.13	26.8%	2.8%	-0.01	2.81		1.34	0
F77	3.31	64.1%	7.1%	0.02	2.81		1.34	1.2
F78	-1.49	0.0%	6.7%	-0.01				1.2
F79	-1.47	37.1%	24.8%	-0.01	1.22			1.2
F80	-1.65	4.7%	12.2%	-0.01				1.2
F81	0.63	7.9%	42.0%	0.00	3.66			1.2
F82	-0.30	23.5%	9.0%	0.00				1.2
F83	0.58	25.1%	9.2%	0.00	1.22			1.2
F84	-0.13	29.9%	7.3%	0.00				1.2
F85	0.59	29.2%	7.4%	0.00	3.66			1.2
F86	-0.67	28.9%	4.0%	0.00		2.81		1.34
F87	0.73	32.3%	0.0%	0.00		2.81		1.34

Table S4. Polymer Ingredients (Organic components). Units are g L<sup>-1</sup>, except formic acid and acetic acid = cm<sup>3</sup> L<sup>-1</sup>. Grey highlight = formulations which dominantly removed Cl<sup>-</sup> ions; Gold highlight = formulations which dominantly removed Na<sup>+</sup> ions.

Trial	Cl <sup>-</sup> Removal	Na <sup>+</sup> Removal	Formic acid	Acetic acid	Malic acid	Citric acid	Tartaric acid	Tea (gallic acid)	Urea
F1	94.9%	95.5%	1						
F2	78.7%	82.7%	1						
F3	94.2%	49.2%	1						
F4	63.7%	85.5%	1						
F5	93.2%	0.0%					0.9		
F6	91.3%	16.4%					0.9		
F7	95.3%	58.9%	1				0.9		
F8	47.2%	0.0%			0.8				
F9	70.1%	45.7%			0.8				
F10	95.5%	77.5%	1		0.8				
F11	94.0%	0.7%				0.83			
F12	94.2%	11.2%				0.83			
F13	91.0%	78.0%	1			0.83			

F14	83.0%	73.9%		0.8	0.83	0.9	
F15	95.8%	83.3%				0.9	
F16	68.6%	74.1%		0.8			
F17	88.5%	93.1%			1.67		
F18	30.7%	80.8%				1.79	
F19	92.8%	38.5%			0.83		
F21	1.1%	62.4%	2				
F23	69.9%	34.2%			0.83		
F24	56.8%	22.4%	1				
F25	71.0%	0.0%		0.8			
F26	60.2%	13.5%	2				
F30	93.9%	53.6%				0.94	
F31	89.5%	73.8%				0.94	
F32	86.7%	77.0%				0.94	
F33	58.7%	0.0%					1.32
F34	18.1%	38.1%					1.32
F35	68.7%	39.0%					1.32
F36	59.2%	21.1%					1.32
F37	51.1%	19.9%					1.32
F48	59.7%	41.0%					1.32
F50	20.9%	14.8%					1.32

Table S5. Observed First Order Rate Constants. Time unit = seconds.

Trial	Change in pH	Cl <sup>-</sup> Removal	Na <sup>+</sup> Removal	PES Change, V	Category	pH Change	Eh Change, mV	Log(Cl <sup>-</sup> Rate Constant)	Log(Na <sup>+</sup> Rate Constant)
F1	3.54	94.9%	95.5%	-0.02	A7d	3.54	-417	-5.46	-5.45
F2	3.92	78.7%	82.7%	0.02	A7f	3.92	-446	-5.75	-5.69
F3	1.00	94.2%	49.2%	0.01	A7e	1.00	-179	-5.48	-6.11
F4	4.23	63.7%	85.5%	0.02	A7d	4.23	-470	-5.93	-5.65
F5	-5.79	93.2%	0.0%	-0.12	A3	-5.79	829	-2.13	
F6	-2.49	91.3%	16.4%	-0.02	A3a	-2.49	-15	-2.17	-3.30
F7	0.68	95.3%	58.9%	0.00	A3b	0.68	133	-5.41	-5.94
F8	-5.27	47.2%	0.0%	-0.09	A4	-5.27	766	-2.75	
F9	-1.78	70.1%	45.7%	-0.02	A4b	-1.78	-10	-2.47	-2.77
F10	1.42	95.5%	77.5%	0.01	A4c	1.42	22	-5.40	-5.72
F11	-4.86	94.0%	0.7%	-0.07	A5	-4.86	684	-2.11	-4.71
F12	-2.12	94.2%	11.2%	-0.02	A5b	-2.12	78	-2.10	-3.48
F13	1.65	91.0%	78.0%	0.01	A5c	1.65	-2	-5.51	-5.71
F14	2.49	83.0%	73.9%	0.01	A3c	2.49	-13	-5.29	-5.41
F15	3.71	95.8%	83.3%	0.02	A3d	3.71	-352	-4.43	-4.68
F16	3.41	68.6%	74.1%	0.02	A4a	3.41	-344	-4.87	-4.81
F17	3.45	88.5%	93.1%	0.02	A5a	3.45	-578	-4.60	-4.51
F18	2.63	30.7%	80.8%	0.01	A3d	2.63	-203	-5.67	-5.02
F19	0.42	92.8%	38.5%	0.00	A5a	0.42	130	-4.52	-5.25
F20	2.96	80.8%	56.2%	0.01	A6f	2.96	-310	-4.72	-5.02
F21	2.55	1.1%	62.4%	0.01	A7g	2.55	-264	-6.88	-4.95
F22	1.64	49.9%	50.0%	0.01	A6g	1.64	-110	-5.10	-5.10
F23	3.56	69.9%	34.2%	0.02	A5a	3.56	-309	-4.86	-5.31
F24	2.36	56.8%	22.4%	0.01	A6i	2.36	-185	-5.01	-5.53
F25	3.30	71.0%	0.0%	0.01	A4a	3.30	-232	-4.84	
F26	2.66	60.2%	13.5%	0.01	A6h	2.66	-181	-4.97	-5.77
F27	2.97	47.3%	2.8%	0.01	A6e	2.97	-185	-5.13	-6.48
F28	3.76	37.7%	33.7%	0.02	A7	3.76	-273	-5.26	-5.32
F29	3.82	46.5%	0.0%	0.02	A7	3.82	-281	-5.14	
F30	-0.72	93.9%	53.6%	-0.01	A7c	-0.72	99	-6.24	-6.80
F31	-0.03	89.5%	73.8%	0.00	A7b	-0.03	27	-6.33	-6.56
F32	0.93	86.7%	77.0%	0.01	A7a	0.93	-71	-6.40	-6.53
F33	-2.62	58.7%	0.0%	-0.03	A2a	-2.62	-18	-2.51	
F34	-2.20	18.1%	38.1%	-0.02	A2a	-2.20	-174	-3.16	-2.78
F35	4.15	68.7%	39.0%	0.02	A2a	4.15	-475	-5.35	-5.72
F36	2.72	59.2%	21.1%	0.01	B1	2.72	-223	-5.28	-5.86
F37	1.94	51.1%	19.9%	0.01	B1	1.94	-178	-5.38	-5.89
F38	-1.90	35.9%	52.2%	-0.02	A6	-1.90	-6	-5.59	-5.37
F39	-1.22	0.0%	34.2%	-0.01	A6	-1.22	-276		-5.32
F40	-1.21	22.9%	36.1%	-0.01	A6b	-1.21	-201	-5.52	-5.29
F41	3.43	47.4%	15.1%	0.02	B2a	3.43	-330	-5.73	-6.32
F42	2.44	49.2%	13.4%	0.01	B2a	2.44	-193	-5.71	-6.38

F43	2.61	49.4%	6.3%	0.01	B2	2.61	-194	-5.10	-6.12
F44	1.36	47.5%	39.5%	0.01	C4	1.36	-28	-5.13	-5.24
F45	1.49	79.3%	30.6%	0.01	A6d	1.49	-151	-5.22	-5.85
F46	3.42	41.8%	56.2%	0.02	A6c	3.42	-604	-5.68	-5.50
F47	-4.58	89.5%	21.8%	-0.04	A1	-4.58	122	-4.88	-5.85
F48	-5.55	59.7%	41.0%	-0.05	A2	-5.55	476	-4.98	-5.21
F49	-3.45	10.2%	8.8%	-0.02	A8	-3.45	711	-5.90	-5.97
F50	-2.10	20.9%	14.8%	-0.01	A2b	-2.10	582	-5.57	-5.73
F51	2.63	99.2%	28.3%	0.01	A8h	2.63	-243	-4.74	-5.89
F52	0.80	88.5%	2.9%	0.00	A8g	0.80	-37	-4.60	-6.47
F53	0.40	34.5%	22.2%	0.00	C1	0.40	22	-5.31	-5.54
F54	-0.38	43.7%	23.3%	0.00	C1	-0.38	11	-5.18	-5.51
F55	0.17	37.6%	43.4%	0.00	C2	0.17	-43	-5.26	-5.18
F56	-3.42	53.4%	14.0%	-0.03	C3	-3.42	367	-5.05	-5.76
F57	3.02	28.1%	38.9%	0.01	C2a	3.02	-257	-5.42	-5.24
F58	-3.35	39.4%	46.8%	-0.04	C3	-3.35	382	-5.24	-5.14
F59	-2.31	58.9%	55.5%	-0.02	C2a	-2.31	282	-4.99	-5.03
F60	-0.17	38.7%	68.3%	0.00	A8i	-0.17	43	-5.25	-4.88
F61	0.81	46.0%	72.0%	0.01	A8i	0.81	-32	-5.15	-4.83
F62	-1.03	23.6%	15.0%	-0.01	D1a	-1.03	110	-5.81	-6.03
F63	-0.93	8.4%	35.3%	-0.01	B3	-0.93	101	-6.29	-5.60
F64	-1.45	22.9%	13.0%	-0.01	D1a	-1.45	124	-6.00	-6.27
F65	4.73	50.3%	5.5%	0.02	B3	4.73	-467	-5.57	-6.66
F66	-0.88	10.4%	1.9%	-0.01	B3a	-0.88	136	-5.89	-6.66
F67	-0.91	3.7%	3.6%	-0.01	D1	-0.91	141	-6.36	-6.38
F68	-3.42	57.3%	0.0%	-0.04	C3	-3.42	370	-5.01	
F69	2.28	66.5%	15.5%	0.01	C2a	2.28	-131	-4.90	-5.71
F70	-3.08	25.0%	11.3%	-0.03	C3	-3.08	304	-5.48	-5.86
F71	3.96	50.9%	25.5%	0.02	C2a	3.96	-329	-5.08	-5.47
F72	-2.34	18.6%	3.1%	-0.02	C3	-2.34	150	-5.62	-6.44
F73	4.24	30.2%	4.1%	0.02	C2a	4.24	-350	-5.38	-6.31
F74	-2.48	0.0%	21.9%	-0.02	A8c	-2.48	-298		-6.14
F75	3.09	58.1%	25.5%	0.01	A8f	3.09	-866	-5.60	-6.07
F76	-1.13	26.8%	2.8%	-0.01	A8b	-1.13	119	-5.44	-6.49
F77	3.31	64.1%	7.1%	0.02	A8e	3.31	-239	-4.93	-6.07
F78	-1.49	0.0%	6.7%	-0.01	A8c	-1.49	-267		-6.10
F79	-1.47	37.1%	24.8%	-0.01	A8d	-1.47	-181	-5.27	-5.48
F80	-1.65	4.7%	12.2%	-0.01	A8c	-1.65	-339	-6.25	-5.82
F81	0.63	7.9%	42.0%	0.00	A8d	0.63	-427	-6.02	-5.20
F82	-0.30	23.5%	9.0%	0.00	C1	-0.30	19	-5.51	-5.96
F83	0.58	25.1%	9.2%	0.00	C4a	0.58	-62	-5.48	-5.95
F84	-0.13	29.9%	7.3%	0.00	C5	-0.13	3	-5.39	-6.06
F85	0.59	29.2%	7.4%	0.00	C4b	0.59	-80	-5.40	-6.05
F86	-0.67	28.9%	4.0%	0.00	D1b	-0.67	38	-5.40	-6.33
F87	0.73	32.3%	0.0%	0.00	A8a	0.73	-108	-5.35	

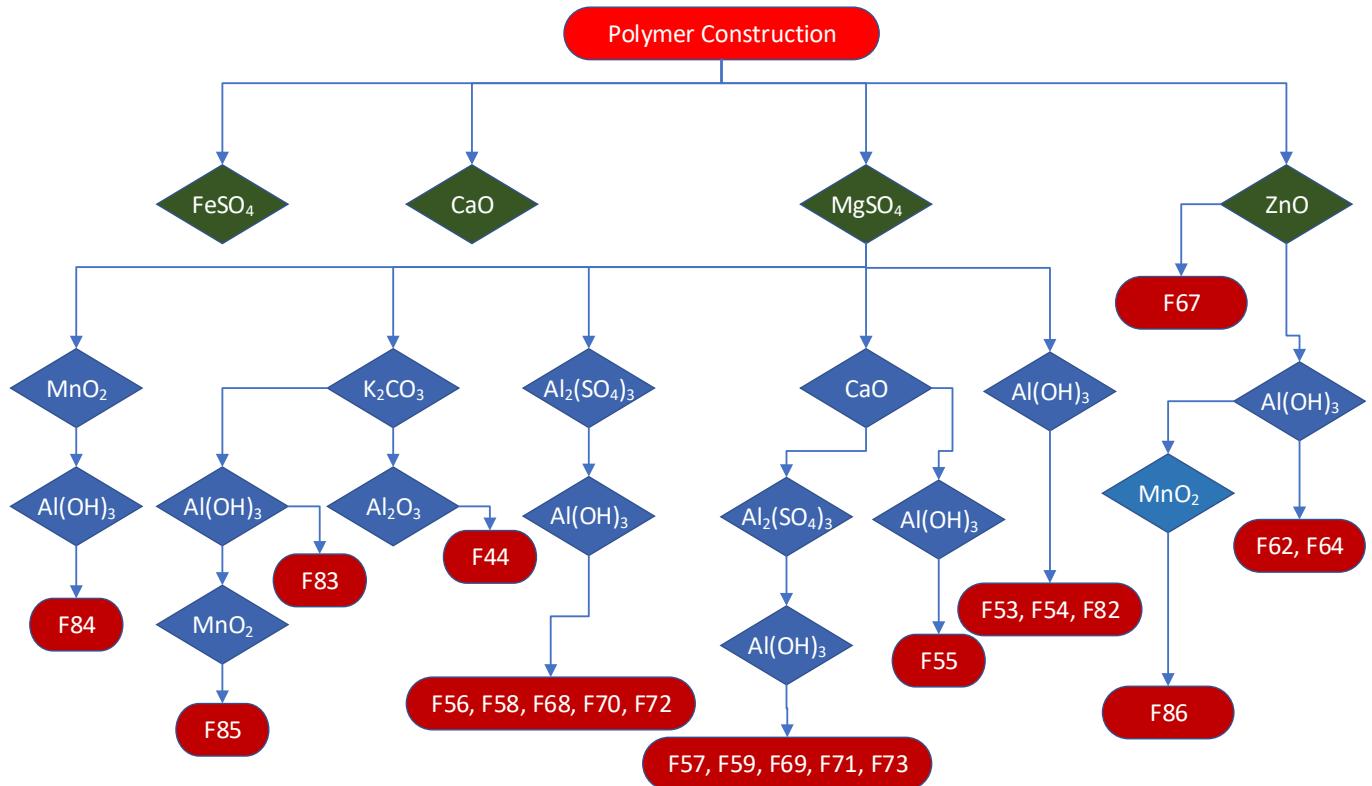


Figure S12. Sol-Gel Polymer Formulation: ZnO + MgSO<sub>4</sub>

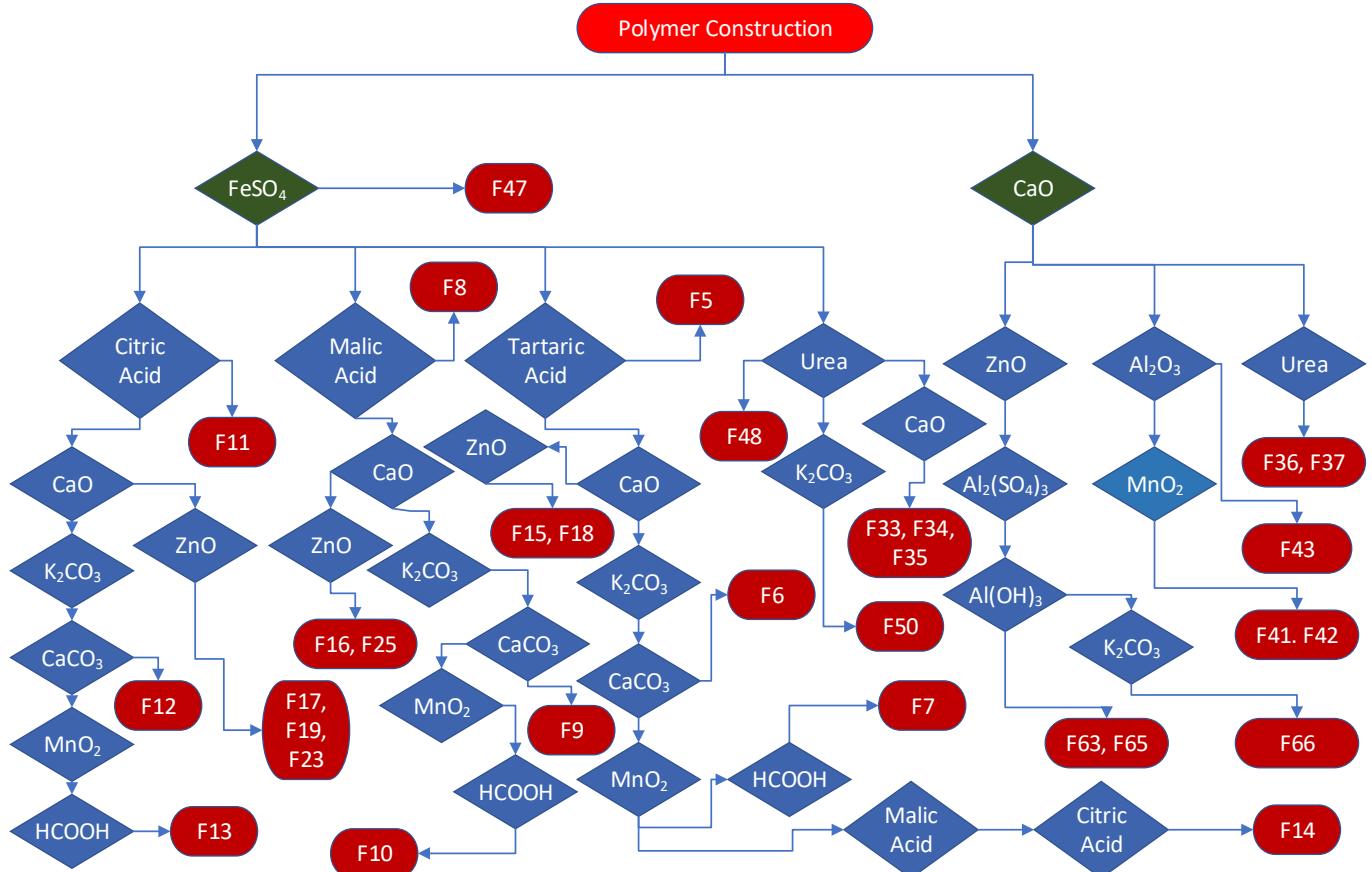


Figure S13. Sol-Gel Polymer Formulation: CaO + FeSO<sub>4</sub>

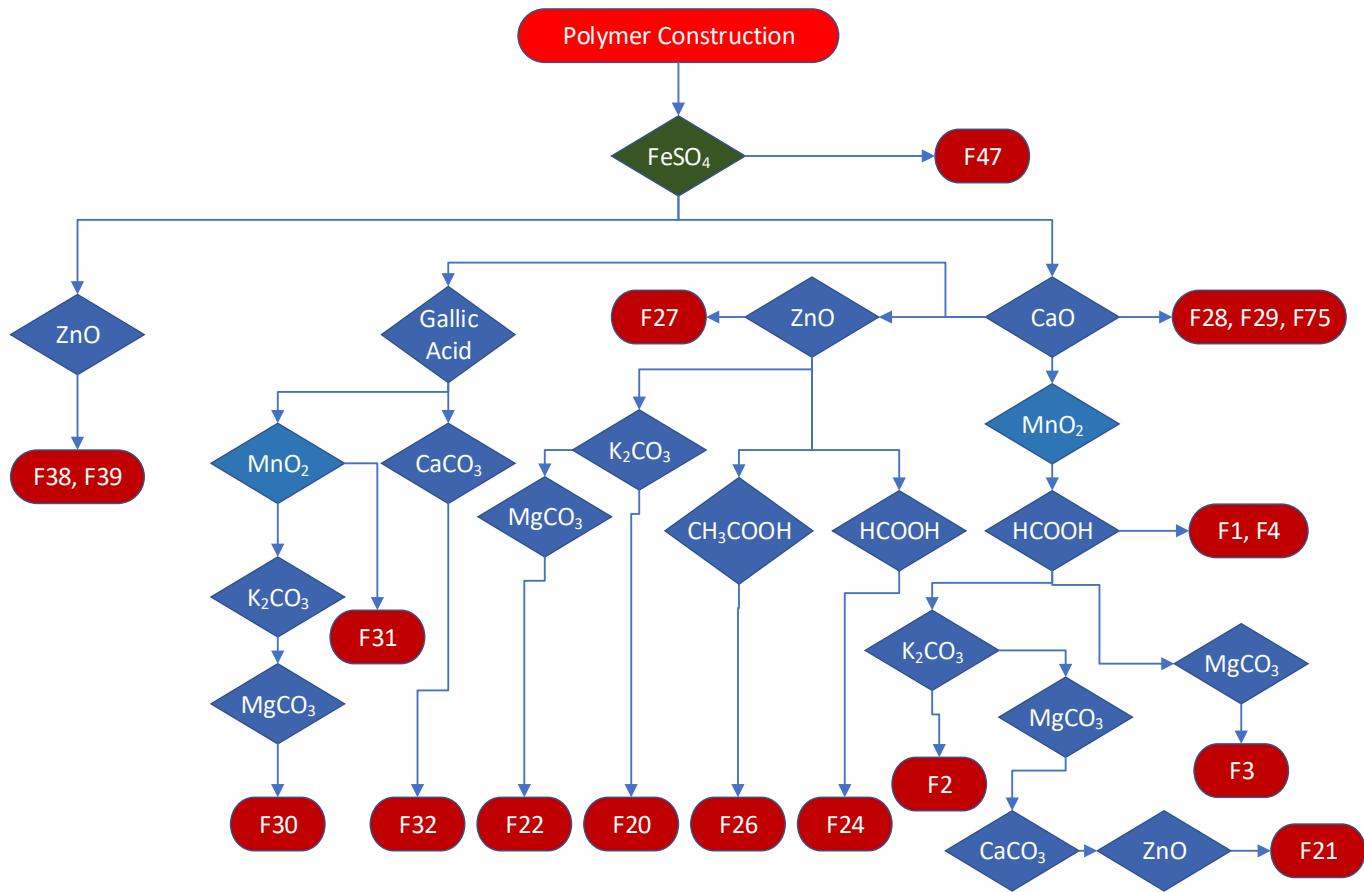


Figure S14. Sol-Gel Polymer Formulation: FeSO<sub>4</sub>@CaO + FeSO<sub>4</sub>@ZnO

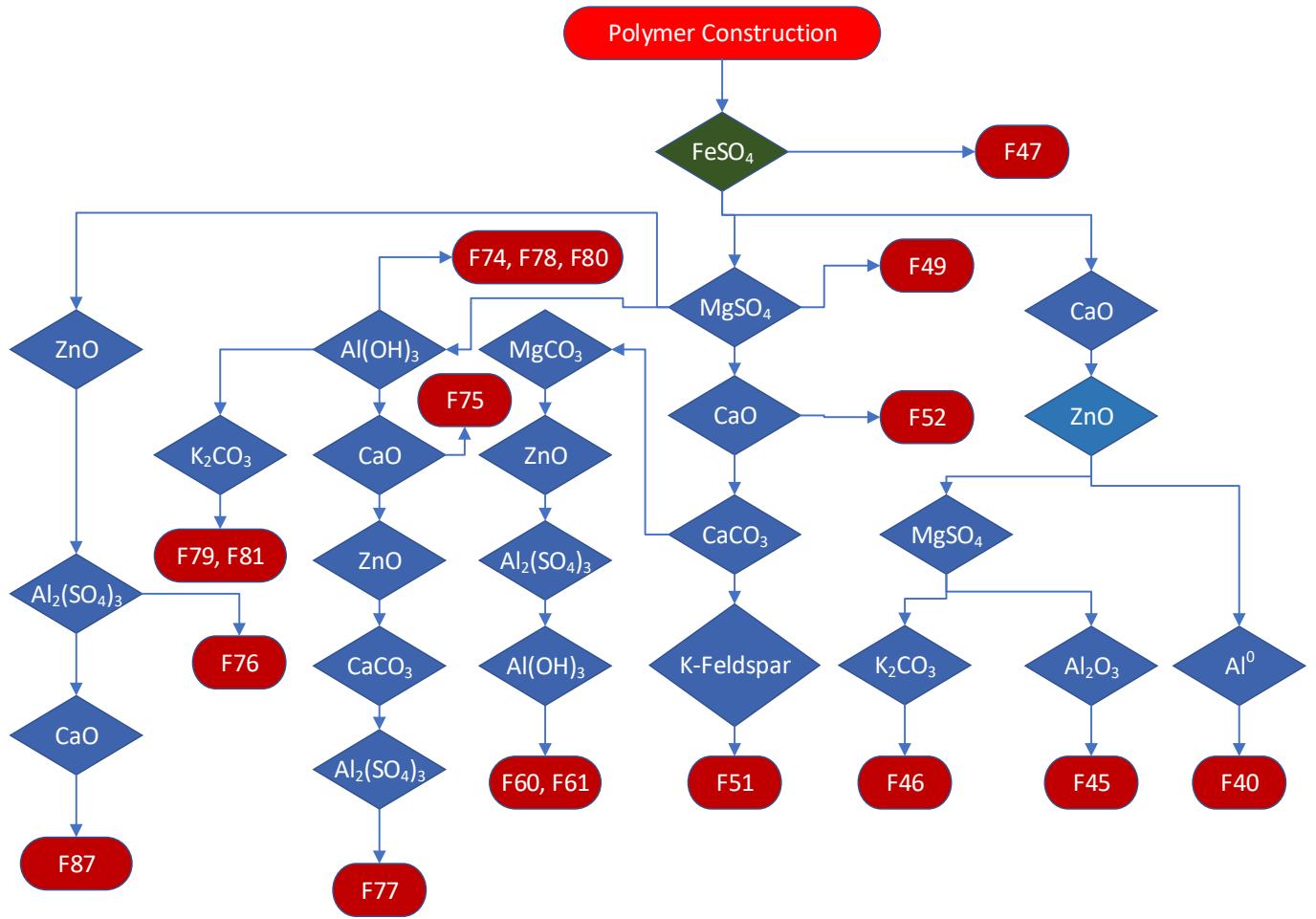


Figure S15. Sol-Gel Polymer Formulation:  $\text{FeSO}_4@\text{CaO} + \text{FeSO}_4@\text{MgSO}_4 + \text{FeSO}_4@\text{ZnO}$

## Supplementary Information Section SC: Crop Yield Analyses

Table S7. Yield decrement constants as a function of salinity, for a number of different crops. Staple global food crops are: Barley, Wheat, Rice, and Corn (Maize). These constants provide indicative, quantitative trends. They will vary with crop variety, water volume provided, water quality, fertiliser provided, pesticide/herbicide strategy, irrigation strategy, agricultural practice, soil, and climate.

Crop	Constants			Crop	Constants		
	a	b	c		a	b	c
Groundnut	1.2076	0.4853	0.3	Almond,			
Lettuce	0.5192	0.0851	0.3	Plum	0.7284	0.1337	0.3
				Apricot	0.9052	0.1748	0.3
				Avocado,			
Onion				Blackberry,			
	0.6352	0.0944	0.3	Boysenberry	0.8898	0.1687	0.3
Peach	0.8244	0.1716	0.3	Barley	0.1	0.4	0.3
Pepper	0.5704	0.1088	0.3	Beans	0.726	0.0855	0.3
Potato	0.4808	0.1023	0.3	Beets	0.3632	0.1828	0.3
Radish	0.5140	0.0759	0.3	Broadbean	0.384	0.0761	0.3
Raspberry	0.8856	0.1073	0.3	Broccoli	0.374	0.1321	0.3
Rice	0.4696	0.1743	0.3	Cabbage	0.3912	0.089	0.3
Safflower	0.4344	0.2874	0.3	Cantaloupe	0.2904	0.0806	0.3

Sesbania	0.2816	0.0812	0.3	Carrot	0.5704	0.0731	0.3
Soybean	0.7984	0.4977	0.3	Cotton	0.1042	0.4046	0.3
Sorghum	0.2820	0.1342	0.3	Corn	0.4808	0.1023	0.3
Spinach	0.3076	0.0779	0.3	Cowpea	0.5548	0.0897	0.3
Strawberry	1.3370	0.1693	0.3	Cucumber	0.5344	0.1691	0.3
Sugar Beet	0.1180	0.4046	0.3	Date Palm	0.1428	0.0707	0.3
Sweet Corn	0.4808	0.1023	0.3	Fig, Olive, Pomegranate	0.3532	0.1187	0.3
Sweet Potato	0.4448	0.0842	0.3	Flax	0.4808	0.1023	0.3
Tomato	0.3992	0.1254	0.3	Grape Grapefruit, Orange, Lemon, Apple, Pear, Walnut	0.3804	0.0699	0.3
Wheat							
	0.1428	0.4286	0.3		0.6452	0.1457	0.3

Table S8. Yield decrement constants as a function of salinity, for grazing and fodder crops. These constants provide indicative, quantitative trends. They will vary with crop variety, water volume provided, water quality, fertiliser provided, pesticide/herbicide strategy, irrigation strategy, agricultural practice, soil, and climate.

Crop	Constants		
	a	b	c
Alfalfa	0.2968	0.0753	0.3
Barley (Hay)	0.0696	0.2143	0.3
Bermuda Grass	0.2568	0.2219	0.3
Clover, berseem	0.7680	0.2201	0.3
Clover, alsike, ladino, red, strawberry	0.4696	0.0863	0.3
Corn (forage)	0.2384	0.0803	0.3
Crested Wheat Grass	0.1600	0.0703	0.3
Harding Grass	0.2988	0.1698	0.3
Lovegrass, Meadow Foxtail	0.3332	0.0834	0.3
Orchard Grass	0.2968	0.0753	0.3
Perennial Rye Grass	0.2984	0.2074	0.3
Sudan Grass	0.1724	0.0602	0.3
Tall Fescue	0.2096	0.1011	0.3
Tall Wheat Grass	0.2748	0.2580	0.3
Trefoil, big	0.7680	0.2201	0.3
Trefoil, Birdsfoot narrow leaf	0.4000	0.2500	0.3
Vetch	0.4440	0.1679	0.3
Wheat Grass	0.1664	0.1548	0.3
Wild Rye, beardless	0.2384	0.0803	0.3

Table S9. Saline feed water used in this study, coupled with the expected yield for crops irrigated with the feed water.  $\text{NaCl} = [\text{Total } \text{Na}^+ \text{ ions} + \text{Total } \text{Cl}^- \text{ ions}]$ . The following assumptions are made. When the feed water is freshwater the expected crop yields are: wheat = 10 t/ha; Cotton = 2 t/ha; Tomato = 25 t/ha; Rice = 10 t/ha; Date = 7 t/ha. Other variants may show a different change in yield with salinity. The yield is calculated where ( $Y_e = Y_s(1 - (a(1+c)S - b))$ ) as follows:  $Y_e = \text{If}(Y_s(1 - (a(1+c)S - b)) < 0, 0, \text{If}(Y_s(1 - (a(1+c)S - b)) > Y_s, Y_s, Y_s(1 - (a(1+c)S - b)))$ .

Trial	Type	NaCl, g/L	Irrigation for Wheat	Useable	Expected	Wheat Yield, t/ha	Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha	Date Yield, t/ha
				for soil water salinity, g/L						
F1	Seawater	43.76	No	56.89	0.00	0.00	0.00	0.00	0.00	0.00
F2	Seawater	43.76	No	56.89	0.00	0.00	0.00	0.00	0.00	0.00
F3	Seawater	43.76	No	56.89	0.00	0.00	0.00	0.00	0.00	0.00
F4	Seawater	43.76	No	56.89	0.00	0.00	0.00	0.00	0.00	0.00
F5	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00	0.00
F6	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00	0.00
F7	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00	0.00
F8	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00	0.00

F9	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00
F10	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00
F11	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00
F12	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00
F13	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00
F14	Seawater	36.54	No	47.50	0.00	0.00	0.00	0.00	0.00
F15	Seawater	43.05	No	55.97	0.00	0.00	0.00	0.00	0.00
F16	Seawater	32.88	No	42.74	0.00	0.00	0.00	0.00	0.00
F17	Seawater	42.23	No	54.90	0.00	0.00	0.00	0.00	0.00
F18	Seawater	45.68	No	59.38	0.00	0.00	0.00	0.00	0.00
F19	Saline water	111.99	No	145.59	0.00	0.00	0.00	0.00	0.00
F20	Saline water	69.25	No	90.03	0.00	0.00	0.00	0.00	0.00
F21	Saline water	49.42	No	64.25	0.00	0.00	0.00	0.00	0.00
F22	Saline water	36.13	No	46.97	0.00	0.00	0.00	0.00	0.00
F23	Saline water	28.41	No	36.93	0.00	0.00	0.00	0.00	0.00
F24	Saline water	16.77	No	21.80	0.00	0.00	0.00	0.00	0.00
F25	Saline water	3.99	Yes	5.19	6.88	1.73	0.00	0.00	2.31
F26	Saline water	5.34	Yes	6.94	4.37	1.36	0.00	0.00	0.56
F27	Saline water	6.19	Marginal	8.05	2.79	1.13	0.00	0.00	0.00
F28	Saline water	14.38	No	18.69	0.00	0.00	0.00	0.00	0.00
F29	Saline water	15.27	No	19.85	0.00	0.00	0.00	0.00	0.00
F30	Seawater	36.44	No	47.37	0.00	0.00	0.00	0.00	0.00
F31	Seawater	36.44	No	47.37	0.00	0.00	0.00	0.00	0.00
F32	Seawater	36.44	No	47.37	0.00	0.00	0.00	0.00	0.00
F33	Seawater	48.52	No	63.08	0.00	0.00	0.00	0.00	0.00
F34	Seawater	48.52	No	63.08	0.00	0.00	0.00	0.00	0.00
F35	Seawater	48.52	No	63.08	0.00	0.00	0.00	0.00	0.00
F36	Saline water	17.33	No	22.53	0.00	0.00	0.00	0.00	0.00
F37	Saline water	4.48	Yes	5.82	5.97	1.60	0.00	0.00	1.67
F38	Saline water	12.45	No	16.19	0.00	0.00	0.00	0.00	0.00
F39	Saline water	40.77	No	53.00	0.00	0.00	0.00	0.00	0.00
F40	Saline water	40.77	No	53.00	0.00	0.00	0.00	0.00	0.00
F41	Saline water	41.80	No	54.34	0.00	0.00	0.00	0.00	0.00
F42	Saline water	24.49	No	31.84	0.00	0.00	0.00	0.00	0.00
F43	Saline water	24.49	No	31.84	0.00	0.00	0.00	0.00	0.00
F44	Saline water	2.74	Yes	3.56	9.20	2.00	0.00	0.00	3.93
F45	Saline water	20.67	No	26.87	0.00	0.00	0.00	0.00	0.00
F46	Saline water	102.61	No	133.39	0.00	0.00	0.00	0.00	0.00
F47	Saline water	9.20	No	11.96	0.00	0.32	0.00	0.00	0.00
F48	Saline water	53.13	No	69.07	0.00	0.00	0.00	0.00	0.00
F49	Saline water	28.31	No	36.80	0.00	0.00	0.00	0.00	0.00
F50	Saline water	28.31	No	36.80	0.00	0.00	0.00	0.00	0.00
F51	Saline water	44.22	No	57.49	0.00	0.00	0.00	0.00	0.00
F52	Saline water	2.18	Yes	2.83	10.00	2.00	0.00	0.00	4.66
F53	Saline water	3.59	Yes	4.67	7.62	1.84	0.00	0.00	2.83
F54	Saline water	34.25	No	44.53	0.00	0.00	0.00	0.00	0.00
F55	Saline water	34.25	No	44.53	0.00	0.00	0.00	0.00	0.00
F56	Saline water	101.40	No	131.82	0.00	0.00	0.00	0.00	0.00
F57	Saline water	101.40	No	131.82	0.00	0.00	0.00	0.00	0.00
F58	Saline water	28.93	No	37.61	0.00	0.00	0.00	0.00	0.00
F59	Saline water	28.93	No	37.61	0.00	0.00	0.00	0.00	0.00
F60	Saline water	2.06	Yes	2.68	10.00	2.00	1.41	0.00	4.82
F61	Saline water	2.06	Yes	2.68	10.00	2.00	1.41	0.00	4.82
F62	Seawater	37.90	No	49.27	0.00	0.00	0.00	0.00	0.00
F63	Seawater	37.90	No	49.27	0.00	0.00	0.00	0.00	0.00
F64	Saline water	30.48	No	39.62	0.00	0.00	0.00	0.00	0.00
F65	Saline water	30.48	No	39.62	0.00	0.00	0.00	0.00	0.00
F66	Saline water	64.71	No	84.12	0.00	0.00	0.00	0.00	0.00
F67	Saline water	64.71	No	84.12	0.00	0.00	0.00	0.00	0.00
F68	Saline water	39.61	No	51.49	0.00	0.00	0.00	0.00	0.00
F69	Saline water	39.61	No	51.49	0.00	0.00	0.00	0.00	0.00
F70	Saline water	30.54	No	39.70	0.00	0.00	0.00	0.00	0.00
F71	Saline water	30.54	No	39.70	0.00	0.00	0.00	0.00	0.00
F72	Saline water	42.83	No	55.68	0.00	0.00	0.00	0.00	0.00

F73	Saline water	42.83	No	55.68	0.00	0.00	0.00	0.00
F74	Saline water	18.49	No	24.04	0.00	0.00	0.00	0.00
F75	Saline water	18.49	No	24.04	0.00	0.00	0.00	0.00
F76	Saline water	10.37	No	13.48	0.00	0.00	0.00	0.00
F77	Saline water	10.37	No	13.48	0.00	0.00	0.00	0.00
F78	Saline water	20.06	No	26.08	0.00	0.00	0.00	0.00
F79	Saline water	20.06	No	26.08	0.00	0.00	0.00	0.00
F80	Saline water	27.09	No	35.22	0.00	0.00	0.00	0.00
F81	Saline water	27.09	No	35.22	0.00	0.00	0.00	0.00
F82	Saline water	17.14	No	22.28	0.00	0.00	0.00	0.00
F83	Saline water	17.14	No	22.28	0.00	0.00	0.00	0.00
F84	Saline water	19.18	No	24.93	0.00	0.00	0.00	0.00
F85	Saline water	19.18	No	24.93	0.00	0.00	0.00	0.00
F86	Saline water	15.70	No	20.41	0.00	0.00	0.00	0.00
F87	Saline water	15.70	No	20.41	0.00	0.00	0.00	0.00

Table S10. Partially desalinated product water produced in this study, coupled with the expected yield for crops irrigated with the product water. NaCl = [Total Na<sup>+</sup> ions + Total Cl<sup>-</sup> ions]. The following assumptions are made. When the feed water is freshwater the expected crop yields are: wheat = 10 t/ha; Cotton = 2 t/ha; Tomato = 25 t/ha;

Rice = 10 t/ha; Date = 7 t/ha. Other variants may show a different change in yield with salinity. The yield is calculated where ( $Y_e = Y_s(1 - (a(1+c)S - b))$ ) as follows:  $Y_e = \text{If}(Y_s(1 - (a(1+c)S - b)) < 0, 0, \text{If}(Y_s(1 - (a(1+c)S - b)) > Y_s, Y_s, Y_s(1 - (a(1+c)S - b)))$ .

Trial	Type	NaCl, g/L	Useable for Irrigation	Expected soil water salinity, g/L	Wheat Yield, t/ha	Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha	Date Yield, t/ha
F1	Seawater	2.13	Yes	2.77	10.00	2.00	0.50	0.00	4.73
F2	Seawater	8.63	Yes	11.22	0.00	0.47	0.00	0.00	0.00
F3	Seawater	10.30	No	13.39	0.00	0.02	0.00	0.00	0.00
F4	Seawater	12.14	No	15.78	0.00	0.00	0.00	0.00	0.00
F5	Seawater	15.94	No	20.72	0.00	0.00	0.00	0.00	0.00
F6	Seawater	14.00	No	18.20	0.00	0.00	0.00	0.00	0.00
F7	Seawater	6.97	Yes	9.06	1.35	0.92	0.00	0.00	0.00
F8	Seawater	26.10	No	33.93	0.00	0.00	0.00	0.00	0.00
F9	Seawater	14.45	No	18.79	0.00	0.00	0.00	0.00	0.00
F10	Seawater	4.25	Yes	5.53	6.40	1.66	0.00	0.00	1.97
F11	Seawater	15.66	No	20.36	0.00	0.00	0.00	0.00	0.00
F12	Seawater	14.11	No	18.34	0.00	0.00	0.00	0.00	0.00
F13	Seawater	5.17	Yes	6.72	4.69	1.41	0.00	0.00	0.78
F14	Seawater	7.53	Yes	9.79	0.31	0.77	0.00	0.00	0.00
F15	Seawater	4.29	Yes	5.58	6.32	1.65	0.00	0.00	1.92
F16	Seawater	9.58	Yes	12.45	0.00	0.21	0.00	0.00	0.00
F17	Seawater	4.12	Yes	5.36	6.64	1.69	0.00	0.00	2.14
F18	Seawater	21.85	No	28.41	0.00	0.00	0.00	0.00	0.00
F19	Saline water	13.73	No	17.85	0.00	0.00	0.00	0.00	0.00
F20	Saline water	18.57	No	24.14	0.00	0.00	0.00	0.00	0.00
F21	Saline water	34.18	No	44.43	0.00	0.00	0.00	0.00	0.00
F22	Saline water	18.08	No	23.50	0.00	0.00	0.00	0.00	0.00
F23	Saline water	11.58	No	15.05	0.00	0.00	0.00	0.00	0.00
F24	Saline water	9.42	Yes	12.25	0.00	0.26	0.00	0.00	0.00
F25	Saline water	2.03	Yes	2.64	10.00	2.00	1.80	0.00	4.86
F26	Saline water	2.99	Yes	3.89	8.74	2.00	0.00	0.00	3.61
F27	Saline water	4.06	Yes	5.28	6.75	1.71	0.00	0.00	2.22
F28	Saline water	9.16	Yes	11.91	0.00	0.33	0.00	0.00	0.00
F29	Saline water	9.63	Yes	12.52	0.00	0.20	0.00	0.00	0.00
F30	Seawater	7.99	Yes	10.39	0.00	0.64	0.00	0.00	0.00
F31	Seawater	6.08	Yes	7.90	3.00	1.16	0.00	0.00	0.00
F32	Seawater	6.22	Yes	8.09	2.74	1.12	0.00	0.00	0.00
F33	Seawater	30.84	No	40.09	0.00	0.00	0.00	0.00	0.00
F34	Seawater	36.05	No	46.87	0.00	0.00	0.00	0.00	0.00

F35	Seawater	20.65	No	26.85	0.00	0.00	0.00	0.00	0.00
F36	Saline water	9.13	Yes	11.87	0.00	0.34	0.00	0.00	0.00
F37	Saline water	2.63	Yes	3.42	9.40	2.00	0.00	0.00	4.08
F38	Saline water	7.28	Yes	9.46	0.77	0.84	0.00	0.00	0.00
F39	Saline water	37.25	No	48.43	0.00	0.00	0.00	0.00	0.00
F40	Saline water	30.07	No	39.09	0.00	0.00	0.00	0.00	0.00
F41	Saline water	27.30	No	35.49	0.00	0.00	0.00	0.00	0.00
F42	Saline water	16.01	No	20.81	0.00	0.00	0.00	0.00	0.00
F43	Saline water	16.70	No	21.71	0.00	0.00	0.00	0.00	0.00
F44	Saline water	1.56	Yes	2.03	10.00	2.00	7.90	2.22	5.47
F45	Saline water	9.20	Yes	11.96	0.00	0.32	0.00	0.00	0.00
F46	Saline water	53.13	No	69.07	0.00	0.00	0.00	0.00	0.00
F47	Saline water	5.71	Yes	7.42	3.69	1.26	0.00	0.00	0.07
F48	Saline water	25.16	No	32.71	0.00	0.00	0.00	0.00	0.00
F49	Saline water	25.65	No	33.35	0.00	0.00	0.00	0.00	0.00
F50	Saline water	23.41	No	30.43	0.00	0.00	0.00	0.00	0.00
F51	Saline water	14.20	No	18.46	0.00	0.00	0.00	0.00	0.00
F52	Saline water	0.85	Yes	1.11	10.00	2.00	17.11	6.55	6.39
F53	Saline water	2.54	Yes	3.30	9.57	2.00	0.00	0.00	4.19
F54	Saline water	22.28	No	28.96	0.00	0.00	0.00	0.00	0.00
F55	Saline water	20.51	No	26.66	0.00	0.00	0.00	0.00	0.00
F56	Saline water	64.63	No	84.02	0.00	0.00	0.00	0.00	0.00
F57	Saline water	68.14	No	88.58	0.00	0.00	0.00	0.00	0.00
F58	Saline water	16.80	No	21.84	0.00	0.00	0.00	0.00	0.00
F59	Saline water	12.21	No	15.87	0.00	0.00	0.00	0.00	0.00
F60	Saline water	1.02	Yes	1.33	10.00	2.00	14.90	5.52	6.17
F61	Saline water	0.90	Yes	1.17	10.00	2.00	16.46	6.25	6.33
F62	Seawater	30.11	No	39.14	0.00	0.00	0.00	0.00	0.00
F63	Seawater	31.16	No	40.51	0.00	0.00	0.00	0.00	0.00
F64	Saline water	24.61	No	31.99	0.00	0.00	0.00	0.00	0.00
F65	Saline water	20.16	No	26.21	0.00	0.00	0.00	0.00	0.00
F66	Saline water	59.28	No	77.06	0.00	0.00	0.00	0.00	0.00
F67	Saline water	62.32	No	81.02	0.00	0.00	0.00	0.00	0.00
F68	Saline water	27.07	No	35.19	0.00	0.00	0.00	0.00	0.00
F69	Saline water	22.31	No	29.00	0.00	0.00	0.00	0.00	0.00
F70	Saline water	25.10	No	32.63	0.00	0.00	0.00	0.00	0.00
F71	Saline water	19.07	No	24.79	0.00	0.00	0.00	0.00	0.00
F72	Saline water	37.14	No	48.28	0.00	0.00	0.00	0.00	0.00
F73	Saline water	33.73	No	43.85	0.00	0.00	0.00	0.00	0.00
F74	Saline water	16.83	Yes	21.88	0.00	0.00	0.00	0.00	0.00
F75	Saline water	10.22	Yes	13.29	0.00	0.04	0.00	0.00	0.00
F76	Saline water	8.54	Yes	11.10	0.00	0.50	0.00	0.00	0.00
F77	Saline water	5.98	Yes	7.77	3.18	1.19	0.00	0.00	0.00
F78	Saline water	19.43	No	25.26	0.00	0.00	0.00	0.00	0.00
F79	Saline water	13.78	No	17.91	0.00	0.00	0.00	0.00	0.00
F80	Saline water	24.64	No	32.03	0.00	0.00	0.00	0.00	0.00
F81	Saline water	19.63	No	25.52	0.00	0.00	0.00	0.00	0.00
F82	Saline water	14.20	No	18.46	0.00	0.00	0.00	0.00	0.00
F83	Saline water	14.04	No	18.25	0.00	0.00	0.00	0.00	0.00
F84	Saline water	15.60	No	20.28	0.00	0.00	0.00	0.00	0.00
F85	Saline water	15.65	No	20.35	0.00	0.00	0.00	0.00	0.00
F86	Saline water	13.09	No	17.02	0.00	0.00	0.00	0.00	0.00
F87	Saline water	13.13	No	17.07	0.00	0.00	0.00	0.00	0.00

Table S11. Increase in crop yield resulting from the use of partially desalinated product water produced in this study. NaCl = [Total Na<sup>+</sup> ions + Total Cl<sup>-</sup> ions]. Calculated using the data in Tables S9 and S10.

Trial	Wheat Yield, t/ha	Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha	Date Yield, t/ha
F1	10.00	2.00	0.50	0.00	4.73
F2	0.00	0.47	0.00	0.00	0.00
F3	0.00	0.02	0.00	0.00	0.00
F4	0.00	0.00	0.00	0.00	0.00

F5	0.00	0.00	0.00	0.00	0.00
F6	0.00	0.00	0.00	0.00	0.00
F7	1.35	0.92	0.00	0.00	0.00
F8	0.00	0.00	0.00	0.00	0.00
F9	0.00	0.00	0.00	0.00	0.00
F10	6.40	1.66	0.00	0.00	1.97
F11	0.00	0.00	0.00	0.00	0.00
F12	0.00	0.00	0.00	0.00	0.00
F13	4.69	1.41	0.00	0.00	0.78
F14	0.31	0.77	0.00	0.00	0.00
F15	6.32	1.65	0.00	0.00	1.92
F16	0.00	0.21	0.00	0.00	0.00
F17	6.64	1.69	0.00	0.00	2.14
F18	0.00	0.00	0.00	0.00	0.00
F19	0.00	0.00	0.00	0.00	0.00
F20	0.00	0.00	0.00	0.00	0.00
F21	0.00	0.00	0.00	0.00	0.00
F22	0.00	0.00	0.00	0.00	0.00
F23	0.00	0.00	0.00	0.00	0.00
F24	0.00	0.26	0.00	0.00	0.00
F25	3.12	0.27	1.80	0.00	2.55
F26	4.36	0.64	0.00	0.00	3.05
F27	3.95	0.58	0.00	0.00	2.22
F28	0.00	0.33	0.00	0.00	0.00
F29	0.00	0.20	0.00	0.00	0.00
F30	0.00	0.64	0.00	0.00	0.00
F31	3.00	1.16	0.00	0.00	0.00
F32	2.74	1.12	0.00	0.00	0.00
F33	0.00	0.00	0.00	0.00	0.00
F34	0.00	0.00	0.00	0.00	0.00
F35	0.00	0.00	0.00	0.00	0.00
F36	0.00	0.34	0.00	0.00	0.00
F37	3.43	0.40	0.00	0.00	2.40
F38	0.77	0.84	0.00	0.00	0.00
F39	0.00	0.00	0.00	0.00	0.00
F40	0.00	0.00	0.00	0.00	0.00
F41	0.00	0.00	0.00	0.00	0.00
F42	0.00	0.00	0.00	0.00	0.00
F43	0.00	0.00	0.00	0.00	0.00
F44	0.80	0.00	7.90	2.22	1.53
F45	0.00	0.32	0.00	0.00	0.00
F46	0.00	0.00	0.00	0.00	0.00
F47	3.69	0.95	0.00	0.00	0.07
F48	0.00	0.00	0.00	0.00	0.00
F49	0.00	0.00	0.00	0.00	0.00
F50	0.00	0.00	0.00	0.00	0.00
F51	0.00	0.00	0.00	0.00	0.00
F52	0.00	0.00	17.11	6.55	1.73
F53	1.95	0.16	0.00	0.00	1.36
F54	0.00	0.00	0.00	0.00	0.00
F55	0.00	0.00	0.00	0.00	0.00
F56	0.00	0.00	0.00	0.00	0.00
F57	0.00	0.00	0.00	0.00	0.00
F58	0.00	0.00	0.00	0.00	0.00
F59	0.00	0.00	0.00	0.00	0.00
F60	0.00	0.00	13.49	5.52	1.35
F61	0.00	0.00	15.05	6.25	1.51
F62	0.00	0.00	0.00	0.00	0.00
F63	0.00	0.00	0.00	0.00	0.00
F64	0.00	0.00	0.00	0.00	0.00
F65	0.00	0.00	0.00	0.00	0.00
F66	0.00	0.00	0.00	0.00	0.00
F67	0.00	0.00	0.00	0.00	0.00
F68	0.00	0.00	0.00	0.00	0.00

F69	0.00	0.00	0.00	0.00	0.00
F70	0.00	0.00	0.00	0.00	0.00
F71	0.00	0.00	0.00	0.00	0.00
F72	0.00	0.00	0.00	0.00	0.00
F73	0.00	0.00	0.00	0.00	0.00
F74	0.00	0.00	0.00	0.00	0.00
F75	0.00	0.04	0.00	0.00	0.00
F76	0.00	0.50	0.00	0.00	0.00
F77	3.18	1.19	0.00	0.00	0.00
F78	0.00	0.00	0.00	0.00	0.00
F79	0.00	0.00	0.00	0.00	0.00
F80	0.00	0.00	0.00	0.00	0.00
F81	0.00	0.00	0.00	0.00	0.00
F82	0.00	0.00	0.00	0.00	0.00
F83	0.00	0.00	0.00	0.00	0.00
F84	0.00	0.00	0.00	0.00	0.00
F85	0.00	0.00	0.00	0.00	0.00
F86	0.00	0.00	0.00	0.00	0.00
F87	0.00	0.00	0.00	0.00	0.00

Table S12. Saline feed water used in this study, coupled with the expected yield for crops irrigated with the feed water. NaCl = Values in Table S6. The following assumptions are made. When the feed water is freshwater the expected crop yields are: wheat = 10 t/ha; Cotton = 2 t/ha; Tomato = 25 t/ha; Rice = 10 t/ha; Date = 7 t/ha. Other variants may show a different change in yield with salinity. The yield is calculated from ( $Y_e = Y_s(1 - (a(1+c)S - b))$ ) as follows:  $Y_e = \text{If}(Y_s(1 - (a(1+c)S - b)) < 0, 0, \text{If}(Y_s(1 - (a(1+c)S - b)) > Y_s, Y_s, Y_s(1 - (a(1+c)S - b)))$ .

Trial	Type	NaCl, g/L	Useable for Irrigation	Expected				
				soil water salinity, g/L	Wheat Yield, t/ha	Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha
F1	Seawater	42.63	No	55.42	0.00	0.00	0.00	0.00
F2	Seawater	42.63	No	55.42	0.00	0.00	0.00	0.00
F3	Seawater	42.63	No	55.42	0.00	0.00	0.00	0.00
F4	Seawater	42.63	No	55.42	0.00	0.00	0.00	0.00
F5	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F6	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F7	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F8	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F9	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F10	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F11	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F12	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F13	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F14	Seawater	35.45	No	46.09	0.00	0.00	0.00	0.00
F15	Seawater	36.70	No	47.71	0.00	0.00	0.00	0.00
F16	Seawater	31.06	No	40.38	0.00	0.00	0.00	0.00
F17	Seawater	41.70	No	54.20	0.00	0.00	0.00	0.00
F18	Seawater	41.19	No	53.55	0.00	0.00	0.00	0.00
F19	Saline water	103.23	No	134.20	0.00	0.00	0.00	0.00
F20	Saline water	63.34	No	82.34	0.00	0.00	0.00	0.00
F21	Saline water	40.76	No	52.99	0.00	0.00	0.00	0.00
F22	Saline water	28.76	No	37.39	0.00	0.00	0.00	0.00
F23	Saline water	25.74	No	33.46	0.00	0.00	0.00	0.00
F24	Saline water	16.62	No	21.61	0.00	0.00	0.00	0.00
F25	Saline water	3.64	Yes	4.74	7.52	1.82	0.00	0.00
F26	Saline water	5.07	Yes	6.59	4.87	1.44	0.00	0.91
F27	Saline water	5.57	Yes	7.25	3.94	1.30	0.00	0.25
F28	Saline water	13.65	No	17.75	0.00	0.00	0.00	0.00
F29	Saline water	13.44	No	17.47	0.00	0.00	0.00	0.00
F30	Seawater	35.50	No	46.15	0.00	0.00	0.00	0.00
F31	Seawater	35.50	No	46.15	0.00	0.00	0.00	0.00

F32	Seawater	35.50	No	46.15	0.00	0.00	0.00	0.00	0.00
F33	Seawater	48.20	No	62.66	0.00	0.00	0.00	0.00	0.00
F34	Seawater	48.20	No	62.66	0.00	0.00	0.00	0.00	0.00
F35	Seawater	48.20	No	62.66	0.00	0.00	0.00	0.00	0.00
F36	Saline water	15.88	No	20.64	0.00	0.00	0.00	0.00	0.00
F37	Saline water	4.11	Yes	5.35	6.65	1.69	0.00	0.00	2.15
F38	Saline water	11.82	No	15.36	0.00	0.00	0.00	0.00	0.00
F39	Saline water	36.08	No	46.90	0.00	0.00	0.00	0.00	0.00
F40	Saline water	36.08	No	46.90	0.00	0.00	0.00	0.00	0.00
F41	Saline water	40.72	No	52.93	0.00	0.00	0.00	0.00	0.00
F42	Saline water	23.05	No	29.97	0.00	0.00	0.00	0.00	0.00
F43	Saline water	23.05	No	29.97	0.00	0.00	0.00	0.00	0.00
F44	Saline water	2.13	Yes	2.77	10.00	2.00	0.53	0.00	4.73
F45	Saline water	16.96	No	22.05	0.00	0.00	0.00	0.00	0.00
F46	Saline water	89.88	No	116.84	0.00	0.00	0.00	0.00	0.00
F47	Saline water	7.43	Yes	9.66	0.48	0.80	0.00	0.00	0.00
F48	Saline water	52.52	No	68.28	0.00	0.00	0.00	0.00	0.00
F49	Saline water	21.70	No	28.21	0.00	0.00	0.00	0.00	0.00
F50	Saline water	21.70	No	28.21	0.00	0.00	0.00	0.00	0.00
F51	Saline water	38.95	No	50.63	0.00	0.00	0.00	0.00	0.00
F52	Saline water	2.01	Yes	2.62	10.00	2.00	2.01	0.00	4.88
F53	Saline water	3.25	Yes	4.23	8.25	1.93	0.00	0.00	3.27
F54	Saline water	30.75	No	39.97	0.00	0.00	0.00	0.00	0.00
F55	Saline water	30.75	No	39.97	0.00	0.00	0.00	0.00	0.00
F56	Saline water	90.24	No	117.31	0.00	0.00	0.00	0.00	0.00
F57	Saline water	90.24	No	117.31	0.00	0.00	0.00	0.00	0.00
F58	Saline water	27.18	No	35.33	0.00	0.00	0.00	0.00	0.00
F59	Saline water	27.18	No	35.33	0.00	0.00	0.00	0.00	0.00
F60	Saline water	1.98	Yes	2.58	10.00	2.00	2.40	0.00	4.92
F61	Saline water	1.98	Yes	2.58	10.00	2.00	2.40	0.00	4.92
F62	Seawater	36.10	No	46.93	0.00	0.00	0.00	0.00	0.00
F63	Seawater	36.10	No	46.93	0.00	0.00	0.00	0.00	0.00
F64	Saline water	29.74	No	38.66	0.00	0.00	0.00	0.00	0.00
F65	Saline water	29.74	No	38.66	0.00	0.00	0.00	0.00	0.00
F66	Saline water	57.06	No	74.18	0.00	0.00	0.00	0.00	0.00
F67	Saline water	57.06	No	74.18	0.00	0.00	0.00	0.00	0.00
F68	Saline water	34.47	No	44.81	0.00	0.00	0.00	0.00	0.00
F69	Saline water	34.47	No	44.81	0.00	0.00	0.00	0.00	0.00
F70	Saline water	24.17	No	31.43	0.00	0.00	0.00	0.00	0.00
F71	Saline water	24.17	No	31.43	0.00	0.00	0.00	0.00	0.00
F72	Saline water	40.54	No	52.70	0.00	0.00	0.00	0.00	0.00
F73	Saline water	40.54	No	52.70	0.00	0.00	0.00	0.00	0.00
F74	Saline water	17.35	No	22.55	0.00	0.00	0.00	0.00	0.00
F75	Saline water	17.35	No	22.55	0.00	0.00	0.00	0.00	0.00
F76	Saline water	10.35	No	13.46	0.00	0.00	0.00	0.00	0.00
F77	Saline water	10.35	No	13.46	0.00	0.00	0.00	0.00	0.00
F78	Saline water	16.82	No	21.87	0.00	0.00	0.00	0.00	0.00
F79	Saline water	16.82	No	21.87	0.00	0.00	0.00	0.00	0.00
F80	Saline water	20.85	No	27.10	0.00	0.00	0.00	0.00	0.00
F81	Saline water	20.85	No	27.10	0.00	0.00	0.00	0.00	0.00
F82	Saline water	15.13	No	19.66	0.00	0.00	0.00	0.00	0.00
F83	Saline water	15.13	No	19.66	0.00	0.00	0.00	0.00	0.00
F84	Saline water	15.62	No	20.31	0.00	0.00	0.00	0.00	0.00
F85	Saline water	15.62	No	20.31	0.00	0.00	0.00	0.00	0.00
F86	Saline water	12.82	No	16.67	0.00	0.00	0.00	0.00	0.00
F87	Saline water	12.82	No	16.67	0.00	0.00	0.00	0.00	0.00

Table S13. Saline product water used in this study, coupled with the expected yield for crops irrigated with the feed water. NaCl = Values in Table S6. The following assumptions are made. When the feed water is freshwater the expected crop yields are: wheat = 10 t/ha; Cotton = 2 t/ha; Tomato = 25 t/ha; Rice = 10 t/ha; Date = 7 t/ha. Other

variants may show a different change in yield with salinity. The yield is calculated from ( $Y_e = Y_s(1 - (a(1+c)S - b))$ ) as follows:  $Y_e = \text{If}(Y_s(1 - (a(1+c)S - b)) < 0, 0, \text{If}(Y_s(1 - (a(1+c)S - b)) > Y_s, Y_s, Y_s(1 - (a(1+c)S - b)))$ .

Trial	Type	NaCl, g/L	Useable for Irrigation	Expected		Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha	Date Yield, t/ha
				soil water salinity, g/L	Wheat Yield, t/ha				
F1	Seawater	2.08	Yes	2.70	10.00	2.00	1.20	0.00	4.80
F2	Seawater	8.17	Yes	10.62	0.00	0.60	0.00	0.00	0.00
F3	Seawater	8.92	Yes	11.60	0.00	0.39	0.00	0.00	0.00
F4	Seawater	10.68	No	13.89	0.00	0.00	0.00	0.00	0.00
F5	Seawater	14.53	No	18.89	0.00	0.00	0.00	0.00	0.00
F6	Seawater	12.26	No	15.94	0.00	0.00	0.00	0.00	0.00
F7	Seawater	6.04	Yes	7.86	3.07	1.17	0.00	0.00	0.00
F8	Seawater	20.29	No	26.37	0.00	0.00	0.00	0.00	0.00
F9	Seawater	11.30	No	14.69	0.00	0.00	0.00	0.00	0.00
F10	Seawater	3.44	Yes	4.47	7.90	1.88	0.00	0.00	3.02
F11	Seawater	14.41	No	18.73	0.00	0.00	0.00	0.00	0.00
F12	Seawater	12.90	No	16.77	0.00	0.00	0.00	0.00	0.00
F13	Seawater	3.95	Yes	5.14	6.95	1.74	0.00	0.00	2.36
F14	Seawater	6.10	Yes	7.93	2.97	1.16	0.00	0.00	0.00
F15	Seawater	3.50	Yes	4.54	7.80	1.86	0.00	0.00	2.95
F16	Seawater	9.27	Yes	12.05	0.00	0.30	0.00	0.00	0.00
F17	Seawater	3.66	Yes	4.76	7.49	1.82	0.00	0.00	2.74
F18	Seawater	19.35	No	25.16	0.00	0.00	0.00	0.00	0.00
F19	Saline water	11.54	No	15.00	0.00	0.00	0.00	0.00	0.00
F20	Saline water	14.94	No	19.42	0.00	0.00	0.00	0.00	0.00
F21	Saline water	30.36	No	39.47	0.00	0.00	0.00	0.00	0.00
F22	Saline water	14.39	No	18.71	0.00	0.00	0.00	0.00	0.00
F23	Saline water	9.59	Yes	12.47	0.00	0.21	0.00	0.00	0.00
F24	Saline water	7.48	Yes	9.73	0.40	0.78	0.00	0.00	0.00
F25	Saline water	1.55	Yes	2.02	10.00	2.00	7.99	2.26	5.48
F26	Saline water	2.35	Yes	3.05	9.92	2.00	0.00	0.00	4.44
F27	Saline water	3.66	Yes	4.76	7.49	1.82	0.00	0.00	2.74
F28	Saline water	8.85	Yes	11.51	0.00	0.41	0.00	0.00	0.00
F29	Saline water	8.93	Yes	11.61	0.00	0.39	0.00	0.00	0.00
F30	Seawater	6.82	Yes	8.86	1.63	0.96	0.00	0.00	0.00
F31	Seawater	4.65	Yes	6.04	5.66	1.55	0.00	0.00	1.45
F32	Seawater	4.91	Yes	6.38	5.17	1.48	0.00	0.00	1.12
F33	Seawater	23.62	No	30.71	0.00	0.00	0.00	0.00	0.00
F34	Seawater	33.13	No	43.07	0.00	0.00	0.00	0.00	0.00
F35	Seawater	16.14	No	20.98	0.00	0.00	0.00	0.00	0.00
F36	Saline water	7.70	Yes	10.01	0.00	0.72	0.00	0.00	0.00
F37	Saline water	2.37	Yes	3.08	9.89	2.00	0.00	0.00	4.42
F38	Saline water	6.53	Yes	8.49	2.16	1.04	0.00	0.00	0.00
F39	Saline water	32.90	No	42.77	0.00	0.00	0.00	0.00	0.00
F40	Saline water	26.46	No	34.40	0.00	0.00	0.00	0.00	0.00
F41	Saline water	21.88	No	28.44	0.00	0.00	0.00	0.00	0.00
F42	Saline water	12.54	No	16.30	0.00	0.00	0.00	0.00	0.00
F43	Saline water	12.93	No	16.81	0.00	0.00	0.00	0.00	0.00
F44	Saline water	1.20	Yes	1.56	10.00	2.00	12.61	4.44	5.94
F45	Saline water	7.43	Yes	9.66	0.48	0.80	0.00	0.00	0.00
F46	Saline water	52.52	No	68.28	0.00	0.00	0.00	0.00	0.00
F47	Saline water	5.49	Yes	7.13	4.10	1.32	0.00	0.00	0.37
F48	Saline water	21.21	No	27.57	0.00	0.00	0.00	0.00	0.00
F49	Saline water	19.65	No	25.54	0.00	0.00	0.00	0.00	0.00
F50	Saline water	17.90	No	23.27	0.00	0.00	0.00	0.00	0.00
F51	Saline water	13.99	No	18.19	0.00	0.00	0.00	0.00	0.00
F52	Saline water	0.71	Yes	0.92	10.00	2.00	18.97	7.43	6.58
F53	Saline water	2.14	Yes	2.78	10.00	2.00	0.36	0.00	4.71
F54	Saline water	17.91	No	23.29	0.00	0.00	0.00	0.00	0.00
F55	Saline water	19.34	No	25.14	0.00	0.00	0.00	0.00	0.00

F56	Saline water	49.59	No	64.46	0.00	0.00	0.00	0.00	0.00
F57	Saline water	66.02	No	85.82	0.00	0.00	0.00	0.00	0.00
F58	Saline water	15.35	No	19.95	0.00	0.00	0.00	0.00	0.00
F59	Saline water	11.73	No	15.24	0.00	0.00	0.00	0.00	0.00
F60	Saline water	0.90	Yes	1.17	10.00	2.00	16.42	6.23	6.32
F61	Saline water	0.80	Yes	1.04	10.00	2.00	17.79	6.88	6.46
F62	Seawater	29.67	No	38.58	0.00	0.00	0.00	0.00	0.00
F63	Seawater	27.82	No	36.17	0.00	0.00	0.00	0.00	0.00
F64	Saline water	23.85	No	31.01	0.00	0.00	0.00	0.00	0.00
F65	Saline water	15.98	No	20.77	0.00	0.00	0.00	0.00	0.00
F66	Saline water	52.50	No	68.25	0.00	0.00	0.00	0.00	0.00
F67	Saline water	54.96	No	71.44	0.00	0.00	0.00	0.00	0.00
F68	Saline water	20.79	No	27.02	0.00	0.00	0.00	0.00	0.00
F69	Saline water	17.21	No	22.37	0.00	0.00	0.00	0.00	0.00
F70	Saline water	19.39	No	25.20	0.00	0.00	0.00	0.00	0.00
F71	Saline water	14.58	No	18.96	0.00	0.00	0.00	0.00	0.00
F72	Saline water	36.99	No	48.09	0.00	0.00	0.00	0.00	0.00
F73	Saline water	31.02	No	40.33	0.00	0.00	0.00	0.00	0.00
F74	Saline water	16.07	No	20.90	0.00	0.00	0.00	0.00	0.00
F75	Saline water	7.95	Yes	10.33	0.00	0.66	0.00	0.00	0.00
F76	Saline water	7.40	Yes	9.61	0.56	0.81	0.00	0.00	0.00
F77	Saline water	4.57	Yes	5.94	5.80	1.57	0.00	0.00	1.55
F78	Saline water	16.71	No	21.73	0.00	0.00	0.00	0.00	0.00
F79	Saline water	10.99	No	14.28	0.00	0.00	0.00	0.00	0.00
F80	Saline water	19.12	No	24.86	0.00	0.00	0.00	0.00	0.00
F81	Saline water	16.71	No	21.73	0.00	0.00	0.00	0.00	0.00
F82	Saline water	11.73	No	15.25	0.00	0.00	0.00	0.00	0.00
F83	Saline water	11.53	No	14.99	0.00	0.00	0.00	0.00	0.00
F84	Saline water	12.06	No	15.67	0.00	0.00	0.00	0.00	0.00
F85	Saline water	12.11	No	15.74	0.00	0.00	0.00	0.00	0.00
F86	Saline water	10.11	No	13.14	0.00	0.07	0.00	0.00	0.00
F87	Saline water	10.07	No	13.09	0.00	0.08	0.00	0.00	0.00

Table S14. Increase in crop yield resulting from the use of partially desalinated product water produced in this study. Calculated using the data in Tables S12 and S13.

Trial	Wheat Yield, t/ha	Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha	Date Yield, t/ha
F1	10.00	2.00	1.20	0.00	4.80
F2	0.00	0.60	0.00	0.00	0.00
F3	0.00	0.39	0.00	0.00	0.00
F4	0.00	0.00	0.00	0.00	0.00
F5	0.00	0.00	0.00	0.00	0.00
F6	0.00	0.00	0.00	0.00	0.00
F7	3.07	1.17	0.00	0.00	0.00
F8	0.00	0.00	0.00	0.00	0.00
F9	0.00	0.00	0.00	0.00	0.00
F10	7.90	1.88	0.00	0.00	3.02
F11	0.00	0.00	0.00	0.00	0.00
F12	0.00	0.00	0.00	0.00	0.00
F13	6.95	1.74	0.00	0.00	2.36
F14	2.97	1.16	0.00	0.00	0.00
F15	7.80	1.86	0.00	0.00	2.95
F16	0.00	0.30	0.00	0.00	0.00
F17	7.49	1.82	0.00	0.00	2.74
F18	0.00	0.00	0.00	0.00	0.00
F19	0.00	0.00	0.00	0.00	0.00
F20	0.00	0.00	0.00	0.00	0.00
F21	0.00	0.00	0.00	0.00	0.00
F22	0.00	0.00	0.00	0.00	0.00
F23	0.00	0.21	0.00	0.00	0.00
F24	0.40	0.78	0.00	0.00	0.00

F25	2.48	0.18	7.99	2.26	2.72
F26	5.05	0.56	0.00	0.00	3.54
F27	3.55	0.52	0.00	0.00	2.49
F28	0.00	0.41	0.00	0.00	0.00
F29	0.00	0.39	0.00	0.00	0.00
F30	1.63	0.96	0.00	0.00	0.00
F31	5.66	1.55	0.00	0.00	1.45
F32	5.17	1.48	0.00	0.00	1.12
F33	0.00	0.00	0.00	0.00	0.00
F34	0.00	0.00	0.00	0.00	0.00
F35	0.00	0.00	0.00	0.00	0.00
F36	0.00	0.72	0.00	0.00	0.00
F37	3.24	0.31	0.00	0.00	2.27
F38	2.16	1.04	0.00	0.00	0.00
F39	0.00	0.00	0.00	0.00	0.00
F40	0.00	0.00	0.00	0.00	0.00
F41	0.00	0.00	0.00	0.00	0.00
F42	0.00	0.00	0.00	0.00	0.00
F43	0.00	0.00	0.00	0.00	0.00
F44	0.00	0.00	12.08	4.44	1.21
F45	0.48	0.80	0.00	0.00	0.00
F46	0.00	0.00	0.00	0.00	0.00
F47	3.62	0.53	0.00	0.00	0.37
F48	0.00	0.00	0.00	0.00	0.00
F49	0.00	0.00	0.00	0.00	0.00
F50	0.00	0.00	0.00	0.00	0.00
F51	0.00	0.00	0.00	0.00	0.00
F52	0.00	0.00	16.96	7.43	1.70
F53	1.75	0.07	0.36	0.00	1.44
F54	0.00	0.00	0.00	0.00	0.00
F55	0.00	0.00	0.00	0.00	0.00
F56	0.00	0.00	0.00	0.00	0.00
F57	0.00	0.00	0.00	0.00	0.00
F58	0.00	0.00	0.00	0.00	0.00
F59	0.00	0.00	0.00	0.00	0.00
F60	0.00	0.00	14.02	6.23	1.40
F61	0.00	0.00	15.39	6.88	1.54
F62	0.00	0.00	0.00	0.00	0.00
F63	0.00	0.00	0.00	0.00	0.00
F64	0.00	0.00	0.00	0.00	0.00
F65	0.00	0.00	0.00	0.00	0.00
F66	0.00	0.00	0.00	0.00	0.00
F67	0.00	0.00	0.00	0.00	0.00
F68	0.00	0.00	0.00	0.00	0.00
F69	0.00	0.00	0.00	0.00	0.00
F70	0.00	0.00	0.00	0.00	0.00
F71	0.00	0.00	0.00	0.00	0.00
F72	0.00	0.00	0.00	0.00	0.00
F73	0.00	0.00	0.00	0.00	0.00
F74	0.00	0.00	0.00	0.00	0.00
F75	0.00	0.66	0.00	0.00	0.00
F76	0.56	0.80	0.00	0.00	0.00
F77	5.80	1.57	0.00	0.00	1.55
F78	0.00	0.00	0.00	0.00	0.00
F79	0.00	0.00	0.00	0.00	0.00
F80	0.00	0.00	0.00	0.00	0.00
F81	0.00	0.00	0.00	0.00	0.00
F82	0.00	0.00	0.00	0.00	0.00
F83	0.00	0.00	0.00	0.00	0.00
F84	0.00	0.00	0.00	0.00	0.00
F85	0.00	0.00	0.00	0.00	0.00
F86	0.00	0.07	0.00	0.00	0.00
F87	0.00	0.08	0.00	0.00	0.00

Table S15. Expected crop yields if the irrigation is with a mixture of 50% freshwater and 50% saline water.

Trial	Type	NaCl, g/L	Useable for Irrigation	Expected soil water salinity, g/L	Wheat Yield, t/ha	Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha	Date Yield, t/ha
F1	Seawater	43.76	No	28.44	0.00	0.00	0.00	0.00	0.00
F2	Seawater	43.76	No	28.44	0.00	0.00	0.00	0.00	0.00
F3	Seawater	43.76	No	28.44	0.00	0.00	0.00	0.00	0.00
F4	Seawater	43.76	No	28.44	0.00	0.00	0.00	0.00	0.00
F5	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F6	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F7	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F8	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F9	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F10	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F11	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F12	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F13	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F14	Seawater	36.54	No	23.75	0.00	0.00	0.00	0.00	0.00
F15	Seawater	43.05	No	27.98	0.00	0.00	0.00	0.00	0.00
F16	Seawater	32.88	No	21.37	0.00	0.00	0.00	0.00	0.00
F17	Seawater	42.23	No	27.45	0.00	0.00	0.00	0.00	0.00
F18	Seawater	45.68	No	29.69	0.00	0.00	0.00	0.00	0.00
F19	Saline water	111.99	No	72.79	0.00	0.00	0.00	0.00	0.00
F20	Saline water	69.25	No	45.01	0.00	0.00	0.00	0.00	0.00
F21	Saline water	49.42	No	32.12	0.00	0.00	0.00	0.00	0.00
F22	Saline water	36.13	No	23.48	0.00	0.00	0.00	0.00	0.00
F23	Saline water	28.41	No	18.47	0.00	0.00	0.00	0.00	0.00
F24	Saline water	16.77	Marginal	10.90	0.00	0.54	0.00	0.00	0.00
F25	Saline water	3.99	Yes	2.59	10.00	2.00	2.25	0.00	4.90
F26	Saline water	5.34	Yes	3.47	9.33	2.00	0.00	0.00	4.03
F27	Saline water	6.19	Yes	4.02	8.54	1.97	0.00	0.00	3.47
F28	Saline water	14.38	Marginal	9.35	0.94	0.86	0.00	0.00	0.00
F29	Saline water	15.27	Marginal	9.93	0.11	0.74	0.00	0.00	0.00
F30	Seawater	36.44	No	23.69	0.00	0.00	0.00	0.00	0.00
F31	Seawater	36.44	No	23.69	0.00	0.00	0.00	0.00	0.00
F32	Seawater	36.44	No	23.69	0.00	0.00	0.00	0.00	0.00
F33	Seawater	48.52	No	31.54	0.00	0.00	0.00	0.00	0.00
F34	Seawater	48.52	No	31.54	0.00	0.00	0.00	0.00	0.00
F35	Seawater	48.52	No	31.54	0.00	0.00	0.00	0.00	0.00
F36	Saline water	17.33	No	11.26	0.00	0.46	0.00	0.00	0.00
F37	Saline water	4.48	Yes	2.91	10.00	2.00	0.00	0.00	4.58
F38	Saline water	12.45	Yes	8.09	2.73	1.12	0.00	0.00	0.00
F39	Saline water	40.77	No	26.50	0.00	0.00	0.00	0.00	0.00
F40	Saline water	40.77	No	26.50	0.00	0.00	0.00	0.00	0.00
F41	Saline water	41.80	No	27.17	0.00	0.00	0.00	0.00	0.00
F42	Saline water	24.49	No	15.92	0.00	0.00	0.00	0.00	0.00
F43	Saline water	24.49	No	15.92	0.00	0.00	0.00	0.00	0.00
F44	Saline water	2.74	Yes	1.78	10.00	2.00	10.36	3.38	5.71
F45	Saline water	20.67	No	13.44	0.00	0.01	0.00	0.00	0.00
F46	Saline water	102.61	No	66.70	0.00	0.00	0.00	0.00	0.00
F47	Saline water	9.20	Yes	5.98	5.75	1.56	0.00	0.00	1.52
F48	Saline water	53.13	No	34.53	0.00	0.00	0.00	0.00	0.00
F49	Saline water	28.31	No	18.40	0.00	0.00	0.00	0.00	0.00
F50	Saline water	28.31	No	18.40	0.00	0.00	0.00	0.00	0.00
F51	Saline water	44.22	No	28.74	0.00	0.00	0.00	0.00	0.00
F52	Saline water	2.18	Yes	1.42	10.00	2.00	13.99	5.09	6.08
F53	Saline water	3.59	Yes	2.33	10.00	2.00	4.85	0.78	5.16
F54	Saline water	34.25	No	22.26	0.00	0.00	0.00	0.00	0.00
F55	Saline water	34.25	No	22.26	0.00	0.00	0.00	0.00	0.00
F56	Saline water	101.40	No	65.91	0.00	0.00	0.00	0.00	0.00
F57	Saline water	101.40	No	65.91	0.00	0.00	0.00	0.00	0.00
F58	Saline water	28.93	No	18.80	0.00	0.00	0.00	0.00	0.00
F59	Saline water	28.93	No	18.80	0.00	0.00	0.00	0.00	0.00
F60	Saline water	2.06	Yes	1.34	10.00	2.00	14.77	5.46	6.16
F61	Saline water	2.06	Yes	1.34	10.00	2.00	14.77	5.46	6.16
F62	Seawater	37.90	No	24.64	0.00	0.00	0.00	0.00	0.00
F63	Seawater	37.90	No	24.64	0.00	0.00	0.00	0.00	0.00

F64	Saline water	30.48	No	19.81	0.00	0.00	0.00	0.00
F65	Saline water	30.48	No	19.81	0.00	0.00	0.00	0.00
F66	Saline water	64.71	No	42.06	0.00	0.00	0.00	0.00
F67	Saline water	64.71	No	42.06	0.00	0.00	0.00	0.00
F68	Saline water	39.61	No	25.75	0.00	0.00	0.00	0.00
F69	Saline water	39.61	No	25.75	0.00	0.00	0.00	0.00
F70	Saline water	30.54	No	19.85	0.00	0.00	0.00	0.00
F71	Saline water	30.54	No	19.85	0.00	0.00	0.00	0.00
F72	Saline water	42.83	No	27.84	0.00	0.00	0.00	0.00
F73	Saline water	42.83	No	27.84	0.00	0.00	0.00	0.00
F74	Saline water	18.49	Marginal	12.02	0.00	0.30	0.00	0.00
F75	Saline water	18.49	Marginal	12.02	0.00	0.30	0.00	0.00
F76	Saline water	10.37	Yes	6.74	4.66	1.40	0.00	0.00
F77	Saline water	10.37	Yes	6.74	4.66	1.40	0.00	0.00
F78	Saline water	20.06	No	13.04	0.00	0.09	0.00	0.00
F79	Saline water	20.06	No	13.04	0.00	0.09	0.00	0.00
F80	Saline water	27.09	No	17.61	0.00	0.00	0.00	0.00
F81	Saline water	27.09	No	17.61	0.00	0.00	0.00	0.00
F82	Saline water	17.14	Marginal	11.14	0.00	0.49	0.00	0.00
F83	Saline water	17.14	Marginal	11.14	0.00	0.49	0.00	0.00
F84	Saline water	19.18	Marginal	12.47	0.00	0.21	0.00	0.00
F85	Saline water	19.18	Marginal	12.47	0.00	0.21	0.00	0.00
F86	Saline water	15.70	Marginal	10.21	0.00	0.68	0.00	0.00
F87	Saline water	15.70	Marginal	10.21	0.00	0.68	0.00	0.00

Table S16. Expected crop yields, if the crop is irrigated with 50% partially desalinated water and 50% freshwater.

Trial	Type	NaCl, g/L	Useable for Irrigation	Expected soil water salinity, g/L	Wheat Yield, t/ha	Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha	Date Yield, t/ha
F1	Seawater	0.78	Yes	0.51	10.00	2.00	23.08	9.36	6.99
F2	Seawater	2.97	Yes	1.93	10.00	2.00	8.87	2.68	5.57
F3	Seawater	8.75	Yes	5.69	6.16	1.62	0.00	0.00	1.81
F4	Seawater	2.50	Yes	1.63	10.00	2.00	11.92	4.11	5.87
F5	Seawater	14.43	Yes	9.38	0.89	0.85	0.00	0.00	0.00
F6	Seawater	12.07	Yes	7.85	3.08	1.17	0.00	0.00	0.00
F7	Seawater	5.93	Yes	3.85	8.78	2.00	0.00	0.00	3.64
F8	Seawater	14.43	Yes	9.38	0.89	0.85	0.00	0.00	0.00
F9	Seawater	7.84	Yes	5.10	7.01	1.75	0.00	0.00	2.40
F10	Seawater	3.25	Yes	2.11	10.00	2.00	7.05	1.82	5.38
F11	Seawater	14.33	Yes	9.31	0.98	0.87	0.00	0.00	0.00
F12	Seawater	12.82	Yes	8.33	2.39	1.07	0.00	0.00	0.00
F13	Seawater	3.18	Yes	2.07	10.00	2.00	7.51	2.04	5.43
F14	Seawater	3.77	Yes	2.45	10.00	2.00	3.68	0.24	5.05
F15	Seawater	3.32	Yes	2.16	10.00	2.00	6.60	1.61	5.34
F16	Seawater	3.46	Yes	2.25	10.00	2.00	5.69	1.18	5.25
F17	Seawater	1.09	Yes	0.71	10.00	2.00	21.06	8.42	6.79
F18	Seawater	3.75	Yes	2.44	10.00	2.00	3.81	0.30	5.06
F19	Saline water	6.46	Yes	4.20	8.29	1.93	0.00	0.00	3.30
F20	Saline water	9.42	Yes	6.12	5.54	1.53	0.00	0.00	1.37
F21	Saline water	9.00	Yes	5.85	5.93	1.59	0.00	0.00	1.65
F22	Saline water	9.37	Yes	6.09	5.59	1.54	0.00	0.00	1.41
F23	Saline water	5.57	Yes	3.62	9.12	2.00	0.00	0.00	3.88
F24	Saline water	4.91	Yes	3.19	9.73	2.00	0.00	0.00	4.30
F25	Saline water	1.23	Yes	0.80	10.00	2.00	20.16	7.99	6.70
F26	Saline water	1.60	Yes	1.04	10.00	2.00	17.76	6.86	6.46
F27	Saline water	1.74	Yes	1.13	10.00	2.00	16.85	6.43	6.36
F28	Saline water	3.30	Yes	2.15	10.00	2.00	6.73	1.67	5.35
F29	Saline water	3.13	Yes	2.03	10.00	2.00	7.83	2.19	5.46
F30	Seawater	6.65	Yes	4.32	8.11	1.91	0.00	0.00	3.17
F31	Seawater	3.76	Yes	2.44	10.00	2.00	3.74	0.27	5.05
F32	Seawater	3.29	Yes	2.14	10.00	2.00	6.79	1.70	5.36
F33	Seawater	18.38	No	11.95	0.00	0.32	0.00	0.00	0.00
F34	Seawater	11.38	Yes	7.40	3.72	1.27	0.00	0.00	0.10
F35	Seawater	11.22	Yes	7.29	3.87	1.29	0.00	0.00	0.20
F36	Saline water	4.27	Yes	2.78	10.00	2.00	0.44	0.00	4.72
F37	Saline water	1.13	Yes	0.73	10.00	2.00	20.80	8.29	6.76
F38	Saline water	2.06	Yes	1.34	10.00	2.00	14.77	5.46	6.16
F39	Saline water	6.78	Yes	4.41	7.99	1.89	0.00	0.00	3.09

F40	Saline water	6.58	Yes	4.28	8.18	1.92	0.00	0.00	3.22
F41	Saline water	13.95	Yes	9.07	1.34	0.92	0.00	0.00	0.00
F42	Saline water	8.65	Yes	5.62	6.26	1.64	0.00	0.00	1.87
F43	Saline water	9.36	Yes	6.08	5.60	1.54	0.00	0.00	1.41
F44	Saline water	0.92	Yes	0.60	10.00	2.00	22.17	8.93	6.90
F45	Saline water	7.01	Yes	4.56	7.78	1.86	0.00	0.00	2.94
F46	Saline water	19.96	No	12.97	0.00	0.11	0.00	0.00	0.00
F47	Saline water	5.48	Yes	3.56	9.20	2.00	0.00	0.00	3.93
F48	Saline water	11.78	Yes	7.66	3.35	1.21	0.00	0.00	0.00
F49	Saline water	15.29	Marginal	9.94	0.09	0.74	0.00	0.00	0.00
F50	Saline water	14.28	Marginal	9.28	1.03	0.87	0.00	0.00	0.00
F51	Saline water	13.99	Yes	9.09	1.30	0.91	0.00	0.00	0.00
F52	Saline water	0.68	Yes	0.44	10.00	2.00	23.72	9.67	7.00
F53	Saline water	1.19	Yes	0.77	10.00	2.00	20.42	8.11	6.72
F54	Saline water	11.31	Yes	7.35	3.79	1.28	0.00	0.00	0.15
F55	Saline water	8.35	Yes	5.43	6.54	1.68	0.00	0.00	2.07
F56	Saline water	37.97	No	24.68	0.00	0.00	0.00	0.00	0.00
F57	Saline water	26.95	No	17.52	0.00	0.00	0.00	0.00	0.00
F58	Saline water	5.19	Yes	3.37	9.47	2.00	0.00	0.00	4.12
F59	Saline water	4.34	Yes	2.82	10.00	2.00	0.00	0.00	4.68
F60	Saline water	0.26	Yes	0.17	10.00	2.00	25.00	10.00	7.00
F61	Saline water	0.23	Yes	0.15	10.00	2.00	25.00	10.00	7.00
F62	Seawater	11.25	Yes	7.31	3.84	1.29	0.00	0.00	0.19
F63	Seawater	8.56	Yes	5.56	6.34	1.65	0.00	0.00	1.93
F64	Saline water	9.73	Yes	6.32	5.25	1.49	0.00	0.00	1.17
F65	Saline water	10.56	Yes	6.86	4.48	1.38	0.00	0.00	0.63
F66	Saline water	15.11	No	9.82	0.26	0.76	0.00	0.00	0.00
F67	Saline water	14.85	No	9.65	0.50	0.80	0.00	0.00	0.00
F68	Saline water	17.74	No	11.53	0.00	0.41	0.00	0.00	0.00
F69	Saline water	14.99	No	9.74	0.37	0.78	0.00	0.00	0.00
F70	Saline water	14.22	Yes	9.24	1.09	0.88	0.00	0.00	0.00
F71	Saline water	11.95	Yes	7.77	3.19	1.19	0.00	0.00	0.00
F72	Saline water	14.28	Yes	9.28	1.03	0.87	0.00	0.00	0.00
F73	Saline water	14.12	Yes	9.18	1.18	0.90	0.00	0.00	0.00
F74	Saline water	5.91	Yes	3.84	8.80	2.00	0.00	0.00	3.65
F75	Saline water	5.64	Yes	3.67	9.05	2.00	0.00	0.00	3.83
F76	Saline water	3.85	Yes	2.50	10.00	2.00	3.16	0.00	4.99
F77	Saline water	3.68	Yes	2.39	10.00	2.00	4.26	0.51	5.10
F78	Saline water	8.84	Yes	5.75	6.08	1.61	0.00	0.00	1.75
F79	Saline water	7.12	Yes	4.63	7.68	1.84	0.00	0.00	2.87
F80	Saline water	13.70	Yes	8.91	1.57	0.95	0.00	0.00	0.00
F81	Saline water	9.06	Yes	5.89	5.88	1.58	0.00	0.00	1.61
F82	Saline water	6.86	Yes	4.46	7.92	1.88	0.00	0.00	3.04
F83	Saline water	6.85	Yes	4.45	7.93	1.88	0.00	0.00	3.04
F84	Saline water	8.81	Yes	5.73	6.11	1.62	0.00	0.00	1.77
F85	Saline water	8.80	Yes	5.72	6.12	1.62	0.00	0.00	1.78
F86	Saline water	7.43	Yes	4.83	7.39	1.80	0.00	0.00	2.67
F87	Saline water	7.74	Yes	5.03	7.10	1.76	0.00	0.00	2.47

Table S17. Increase in crop yield when the crop is irrigated with 50% fresh water and 50% partially desalinated water. Calculated by subtracting the results in Table S15 from the results in Table S16.

Trial	Type	Wheat Yield, t/ha	Cotton Yield, t/ha	Tomato Yield, t/ha	Rice Yield, t/ha	Date Yield, t/ha
F1	Seawater	10.00	2.00	23.08	9.36	6.99
F2	Seawater	10.00	2.00	8.87	2.68	5.57
F3	Seawater	6.16	1.62	0.00	0.00	1.81
F4	Seawater	10.00	2.00	11.92	4.11	5.87
F5	Seawater	0.89	0.85	0.00	0.00	0.00
F6	Seawater	3.08	1.17	0.00	0.00	0.00
F7	Seawater	8.78	2.00	0.00	0.00	3.64
F8	Seawater	0.89	0.85	0.00	0.00	0.00
F9	Seawater	7.01	1.75	0.00	0.00	2.40
F10	Seawater	10.00	2.00	7.05	1.82	5.38
F11	Seawater	0.98	0.87	0.00	0.00	0.00
F12	Seawater	2.39	1.07	0.00	0.00	0.00
F13	Seawater	10.00	2.00	7.51	2.04	5.43
F14	Seawater	10.00	2.00	3.68	0.24	5.05

F15	Seawater	10.00	2.00	6.60	1.61	5.34
F16	Seawater	10.00	2.00	5.69	1.18	5.25
F17	Seawater	10.00	2.00	21.06	8.42	6.79
F18	Seawater	10.00	2.00	3.81	0.30	5.06
F19	Saline water	8.29	1.93	0.00	0.00	3.30
F20	Saline water	5.54	1.53	0.00	0.00	1.37
F21	Saline water	5.93	1.59	0.00	0.00	1.65
F22	Saline water	5.59	1.54	0.00	0.00	1.41
F23	Saline water	9.12	2.00	0.00	0.00	3.88
F24	Saline water	9.73	1.46	0.00	0.00	4.30
F25	Saline water	0.00	0.00	17.90	7.99	1.79
F26	Saline water	0.67	0.00	17.76	6.86	2.43
F27	Saline water	1.46	0.03	16.85	6.43	2.89
F28	Saline water	9.06	1.14	6.73	1.67	5.35
F29	Saline water	9.89	1.26	7.83	2.19	5.46
F30	Seawater	8.11	1.91	0.00	0.00	3.17
F31	Seawater	10.00	2.00	3.74	0.27	5.05
F32	Seawater	10.00	2.00	6.79	1.70	5.36
F33	Seawater	0.00	0.32	0.00	0.00	0.00
F34	Seawater	3.72	1.27	0.00	0.00	0.10
F35	Seawater	3.87	1.29	0.00	0.00	0.20
F36	Saline water	10.00	1.54	0.44	0.00	4.72
F37	Saline water	0.00	0.00	20.80	8.29	2.18
F38	Saline water	7.27	0.88	14.77	5.46	6.16
F39	Saline water	7.99	1.89	0.00	0.00	3.09
F40	Saline water	8.18	1.92	0.00	0.00	3.22
F41	Saline water	1.34	0.92	0.00	0.00	0.00
F42	Saline water	6.26	1.64	0.00	0.00	1.87
F43	Saline water	5.60	1.54	0.00	0.00	1.41
F44	Saline water	0.00	0.00	11.81	5.56	1.18
F45	Saline water	7.78	1.85	0.00	0.00	2.94
F46	Saline water	0.00	0.11	0.00	0.00	0.00
F47	Saline water	3.45	0.44	0.00	0.00	2.42
F48	Saline water	3.35	1.21	0.00	0.00	0.00
F49	Saline water	0.09	0.74	0.00	0.00	0.00
F50	Saline water	1.03	0.87	0.00	0.00	0.00
F51	Saline water	1.30	0.91	0.00	0.00	0.00
F52	Saline water	0.00	0.00	9.73	4.58	0.92
F53	Saline water	0.00	0.00	15.57	7.33	1.56
F54	Saline water	3.79	1.28	0.00	0.00	0.15
F55	Saline water	6.54	1.68	0.00	0.00	2.07
F56	Saline water	0.00	0.00	0.00	0.00	0.00
F57	Saline water	0.00	0.00	0.00	0.00	0.00
F58	Saline water	9.47	2.00	0.00	0.00	4.12
F59	Saline water	10.00	2.00	0.00	0.00	4.68
F60	Saline water	0.00	0.00	10.23	4.54	0.84
F61	Saline water	0.00	0.00	10.23	4.54	0.84
F62	Seawater	3.84	1.29	0.00	0.00	0.19
F63	Seawater	6.34	1.65	0.00	0.00	1.93
F64	Saline water	5.25	1.49	0.00	0.00	1.17
F65	Saline water	4.48	1.38	0.00	0.00	0.63
F66	Saline water	0.26	0.76	0.00	0.00	0.00
F67	Saline water	0.50	0.80	0.00	0.00	0.00
F68	Saline water	0.00	0.41	0.00	0.00	0.00
F69	Saline water	0.37	0.78	0.00	0.00	0.00
F70	Saline water	1.09	0.88	0.00	0.00	0.00
F71	Saline water	3.19	1.19	0.00	0.00	0.00
F72	Saline water	1.03	0.87	0.00	0.00	0.00
F73	Saline water	1.18	0.90	0.00	0.00	0.00
F74	Saline water	8.80	1.70	0.00	0.00	3.65
F75	Saline water	9.05	1.70	0.00	0.00	3.83
F76	Saline water	5.34	0.60	3.16	0.00	4.24
F77	Saline water	5.34	0.60	4.26	0.51	4.35
F78	Saline water	6.08	1.52	0.00	0.00	1.75
F79	Saline water	7.68	1.75	0.00	0.00	2.87
F80	Saline water	1.57	0.95	0.00	0.00	0.00
F81	Saline water	5.88	1.58	0.00	0.00	1.61
F82	Saline water	7.92	1.39	0.00	0.00	3.04
F83	Saline water	7.93	1.39	0.00	0.00	3.04
F84	Saline water	6.11	1.40	0.00	0.00	1.77
F85	Saline water	6.12	1.41	0.00	0.00	1.78

F86	Saline water	7.39	1.12	0.00	0.00	2.67
F87	Saline water	7.10	1.08	0.00	0.00	2.47

## Supplementary Information Section SD: Polymer Categories

### SD1 Category A: n-Fe(a,b,c) polymers and variants

- a. Category A1: n-Fe(a,b,c); (Average Cl<sup>-</sup> Removal = 89.5%; Average Na<sup>+</sup> Removal = 21.93%; Average Observation Period = 24 h); Fe<sup>0</sup>:n-Fe(a,b,c), and n-Fe<sup>0</sup>:n-Fe(a,b,c) have previously been used to desalinate water.
- b. Category A2: n-Fe(a,b,c)@Urea polymer; (Average Cl<sup>-</sup> Removal = 59.66%; Average Na<sup>+</sup> Removal = 40.98%; Average Observation Period = 24 h); Fe<sup>0</sup>:n-Fe(a,b,c)@Urea polymer and SiO<sub>2</sub>@n-Fe(a,b,c)@Urea polymer have been used to catalytically desalinate water with a single batch of polymer being used to sequentially process (desalinate) more than 70 batches of water and more than 43 m<sup>3</sup> water g<sup>-1</sup> Fe.
  - i. Category A2a: n-Fe(a,b,c)@Urea polymer@Ca(OH)<sub>2</sub>; (Average Cl<sup>-</sup> Removal = 48.51%; Average Na<sup>+</sup> Removal = 25.68%; Average Observation Period = 24 h);
  - ii. Category A2b: n-Fe(a,b,c)@Urea polymer@K<sub>2</sub>CO<sub>3</sub>; (Average Cl<sup>-</sup> Removal = 20.88%; Average Na<sup>+</sup> Removal = 14.85%; Average Observation Period = 24 h);
- c. Category A3: n-Fe(a,b,c)@Tartaric Acid polymer; (Average Cl<sup>-</sup> Removal = 93.17%; Average Na<sup>+</sup> Removal = 0.0%; Average Observation Period = 0.1 h);
  - i. Category A3a: n-Fe(a,b,c)@Tartaric Acid@Ca(OH)<sub>2</sub>@K<sub>2</sub>CO<sub>3</sub>@CaCO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 91.27%; Average Na<sup>+</sup> Removal = 16.35%; Average Observation Period = 0.1 h);
  - ii. Category A3b: n-Fe(a,b,c)@Tartaric Acid@Ca(OH)<sub>2</sub> @K<sub>2</sub>CO<sub>3</sub>@CaCO<sub>3</sub>@MnO<sub>2</sub>@HCOOH polymer; (Average Cl<sup>-</sup> Removal = 95.3%; Average Na<sup>+</sup> Removal = 58.91%; Average Observation Period = 216 h);
  - iii. Category A3c: n-Fe(a,b,c)@Tartaric Acid@Ca(OH)<sub>2</sub> @K<sub>2</sub>CO<sub>3</sub>@CaCO<sub>3</sub>@MnO<sub>2</sub>@citric acid@malic acid polymer; (Average Cl<sup>-</sup> Removal = 82.99%; Average Na<sup>+</sup> Removal = 73.87%; Average Observation Period = 96 h);
  - iv. Category A3d: n-Fe(a,b,c)@Tartaric Acid@Ca(OH)<sub>2</sub> @Zn(OH)<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 62.35%; Average Na<sup>+</sup> Removal = 82.05%; Average Observation Period = 36 h);
- d. Category A4: n-Fe(a,b,c)@Malic Acid polymer; (Average Cl<sup>-</sup> Removal = 47.22%; Average Na<sup>+</sup> Removal = 0.0%; Average Observation Period = 0.1 h);
  - i. Category A4a: n-Fe(a,b,c)@Malic Acid@Ca(OH)<sub>2</sub> @Zn(OH)<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 69.83%; Average Na<sup>+</sup> Removal = 37.05%; Average Observation Period = 24 h);
  - ii. Category A4b: n-Fe(a,b,c)@Malic Acid@Ca(OH)<sub>2</sub>@K<sub>2</sub>CO<sub>3</sub>@CaCO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 70.1%; Average Na<sup>+</sup> Removal = 45.67%; Average Observation Period = 0.1 h);
  - iii. Category A4c: n-Fe(a,b,c)@Malic Acid@Ca(OH)<sub>2</sub> @K<sub>2</sub>CO<sub>3</sub>@CaCO<sub>3</sub>@MnO<sub>2</sub>@HCOOH polymer; (Average Cl<sup>-</sup> Removal = 77.48%; Average Na<sup>+</sup> Removal = 88.37%; Average Observation Period = 216 h);
- e. Category A5: n-Fe(a,b,c)@Citric Acid polymer; (Average Cl<sup>-</sup> Removal = 93.98%; Average Na<sup>+</sup> Removal = 0.69%; Average Observation Period = 0.1 h);
  - i. Category A5a: n-Fe(a,b,c)@Citric Acid@Ca(OH)<sub>2</sub> @Zn(OH)<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 55.29%; Average Na<sup>+</sup> Removal = 79.07%; Average Observation Period = 24 h);
  - ii. Category A5b: n-Fe(a,b,c)@Citric Acid@Ca(OH)<sub>2</sub>@K<sub>2</sub>CO<sub>3</sub>@CaCO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 94.17%; Average Na<sup>+</sup> Removal = 11.16%; Average Observation Period = 0.1 h);
  - iii. Category A5c: n-Fe(a,b,c)@Citric Acid@Ca(OH)<sub>2</sub> @K<sub>2</sub>CO<sub>3</sub>@CaCO<sub>3</sub>@MnO<sub>2</sub>@HCOOH polymer; (Average Cl<sup>-</sup> Removal = 91.00%; Average Na<sup>+</sup> Removal = 77.96%; Average Observation Period = 216 h);

- f. Category A6: n-Fe(a,b,c)@ZnO polymer; (Average Cl<sup>-</sup> Removal = 17.94%; Average Na<sup>+</sup> Removal = 43.19%; Average Observation Period = 36 h);
- i. Category A6a: n-Fe(a,b,c)@ZnO@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>@Ca(OH)<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 32.20%; Average Na<sup>+</sup> Removal = 0.00%; Average Observation Period = 24 h);
  - ii. Category A6b: n-Fe(a,b,c)@CaO@ZnO@Al<sup>0</sup> polymer; (Average Cl<sup>-</sup> Removal = 22.91%; Average Na<sup>+</sup> Removal = 36.12%; Average Observation Period = 24 h);
  - iii. Category A6c: n-Fe(a,b,c)@CaO@ZnO@MgSO<sub>4</sub>@K<sub>2</sub>CO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 41.85%; Average Na<sup>+</sup> Removal = 56.2%; Average Observation Period = 72 h);
  - iv. Category A6d: n-Fe(a,b,c)@CaO@ZnO@MgSO<sub>4</sub>@Al<sub>2</sub>O<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 79.28%; Average Na<sup>+</sup> Removal = 30.59%; Average Observation Period = 72 h);
  - v. Category A6e: n-Fe(a,b,c)@CaO@ZnO polymer; (Average Cl<sup>-</sup> Removal = 47.27%; Average Na<sup>+</sup> Removal = 2.79%; Average Observation Period = 24 h);
  - vi. Category A6f: n-Fe(a,b,c)@CaO@ZnO@K<sub>2</sub>CO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 80.84%; Average Na<sup>+</sup> Removal = 56.19%; Average Observation Period = 24 h);
  - vii. Category A6g: n-Fe(a,b,c)@CaO@ZnO@K<sub>2</sub>CO<sub>3</sub>@MgCO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 49.94%; Average Na<sup>+</sup> Removal = 49.96%; Average Observation Period = 24 h);
  - viii. Category A6h: n-Fe(a,b,c)@CaO@ZnO@CH<sub>3</sub>COOH polymer; (Average Cl<sup>-</sup> Removal = 60.17%; Average Na<sup>+</sup> Removal = 13.51%; Average Observation Period = 24 h);
  - ix. Category A6i: n-Fe(a,b,c)@CaO@ZnO@HCOOH polymer; (Average Cl<sup>-</sup> Removal = 56.8%; Average Na<sup>+</sup> Removal = 22.43%; Average Observation Period = 24 h);
- g. Category A7: n-Fe(a,b,c)@CaO polymer; (Average Cl<sup>-</sup> Removal = 47.39%; Average Na<sup>+</sup> Removal = 19.74%; Average Observation Period = 48 h);
- i. Category A7a: n-Fe(a,b,c)@CaO@Gallic acid@CaCO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 86.75%; Average Na<sup>+</sup> Removal = 77.04%; Average Observation Period = 1398 h);
  - ii. Category A7b: n-Fe(a,b,c)@CaO@Gallic acid@MnO<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 89.51%; Average Na<sup>+</sup> Removal = 73.76%; Average Observation Period = 1344 h);
  - iii. Category A7c: n-Fe(a,b,c)@CaO@Gallic acid@MnO<sub>2</sub>@K<sub>2</sub>CO<sub>3</sub>@MgCO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 93.94%; Average Na<sup>+</sup> Removal = 53.59%; Average Observation Period = 1344 h);
  - iv. Category A7d: n-Fe(a,b,c)@CaO@MnO<sub>2</sub>@HCOOH polymer; (Average Cl<sup>-</sup> Removal = 79.31%; Average Na<sup>+</sup> Removal = 90.47%; Average Observation Period = 241 h);
  - v. Category A7e: n-Fe(a,b,c)@CaO@MnO<sub>2</sub>@HCOOH @MgCO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 94.16%; Average Na<sup>+</sup> Removal = 49.16%; Average Observation Period = 241 h);
  - vi. Category A7f: n-Fe(a,b,c)@CaO@MnO<sub>2</sub>@HCOOH@K<sub>2</sub>CO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 78.68%; Average Na<sup>+</sup> Removal = 82.74%; Average Observation Period = 241 h);
  - vii. Category A7g: n-Fe(a,b,c)@CaO@MnO<sub>2</sub>@HCOOH@K<sub>2</sub>CO<sub>3</sub> @MgCO<sub>3</sub>@CaCO<sub>3</sub>@ZnO polymer; (Average Cl<sup>-</sup> Removal = 1.14%; Average Na<sup>+</sup> Removal = 62.42%; Average Observation Period = 24 h);
- h. Category A8: n-Fe(a,b,c)@MgSO<sub>4</sub> polymer; (Average Cl<sup>-</sup> Removal = 10.23%; Average Na<sup>+</sup> Removal = 8.83%; Average Observation Period = 24 h);
- i. Category A8a: n-Fe(a,b,c)@MgSO<sub>4</sub>@ZnO@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>@CaO polymer; (Average Cl<sup>-</sup> Removal = 32.29%; Average Na<sup>+</sup> Removal = 0.00%; Average Observation Period = 24 h);
  - ii. Category A8b: n-Fe(a,b,c)@MgSO<sub>4</sub>@ZnO@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 26.83%; Average Na<sup>+</sup> Removal = 2.78%; Average Observation Period = 24 h);
  - iii. Category A8c: n-Fe(a,b,c)@MgSO<sub>4</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 1.57%; Average Na<sup>+</sup> Removal = 13.61%; Average Observation Period = 48 h);
  - iv. Category A8d: n-Fe(a,b,c)@MgSO<sub>4</sub>@Al(OH)<sub>3</sub>@K<sub>2</sub>CO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 22.52%; Average Na<sup>+</sup> Removal = 33.39%; Average Observation Period = 24 h);
  - v. Category A8e: n-Fe(a,b,c)@MgSO<sub>4</sub>@Al(OH)<sub>3</sub>@CaO @CaCO<sub>3</sub>@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 64.12%; Average Na<sup>+</sup> Removal = 7.07%; Average Observation Period = 24 h);

- vi. Category A8f: n-Fe(a,b,c)@MgSO<sub>4</sub>@Al(OH)<sub>3</sub>@CaO polymer; (Average Cl<sup>-</sup> Removal = 58.06%; Average Na<sup>+</sup> Removal = 25.50%; Average Observation Period = 96 h);
- vii. Category A8g: n-Fe(a,b,c)@MgSO<sub>4</sub>@CaO polymer; (Average Cl<sup>-</sup> Removal = 88.51%; Average Na<sup>+</sup> Removal = 2.86%; Average Observation Period = 24 h);
- viii. Category A8h: n-Fe(a,b,c)@MgSO<sub>4</sub>@CaO@CaCO<sub>3</sub>@K-feldspar polymer; (Average Cl<sup>-</sup> Removal = 99.15%; Average Na<sup>+</sup> Removal = 28.26%; Average Observation Period = 24 h);
- ix. Category A8i: n-Fe(a,b,c)@MgSO<sub>4</sub>@CaO@CaCO<sub>3</sub>@MgCO<sub>3</sub> @ZnO@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 42.34%; Average Na<sup>+</sup> Removal = 70.12%; Average Observation Period = 24 h);

### **SD2 Category B: n-Ca(OH)<sub>2</sub> polymers**

- a. Category B1: n-Ca(OH)<sub>2</sub>@Urea polymer; (Average Cl<sup>-</sup> Removal = 55.18%; Average Na<sup>+</sup> Removal = 20.46%; Average Observation Period = 48 h);
- b. Category B2: n-Ca(OH)<sub>2</sub>@Al<sub>2</sub>O<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 49.38%; Average Na<sup>+</sup> Removal = 6.31%; Average Observation Period = 24 h);
  - i. Category B2a: n-Ca(OH)<sub>2</sub>@Al<sub>2</sub>O<sub>3</sub>@MnO<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 48.3%; Average Na<sup>+</sup> Removal = 14.28%; Average Observation Period = 96 h);
- c. Category B3: n-Ca(OH)<sub>2</sub>@ZnO@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 29.33%; Average Na<sup>+</sup> Removal = 20.43%; Average Observation Period = 60 h);
  - a. Category B3a: n-Ca(OH)<sub>2</sub>@ZnO@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>@Al(OH)<sub>3</sub>@K<sub>2</sub>CO<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 10.42%; Average Na<sup>+</sup> Removal = 1.88%; Average Observation Period = 24 h)

### **SD3 Category C: n-Mg(OH)<sub>2</sub> polymers**

- a. Category C1: n-Mg(OH)<sub>2</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 33.92%; Average Na<sup>+</sup> Removal = 18.19%; Average Observation Period = 24 h);
- b. Category C2: n-Mg(OH)<sub>2</sub>@Ca(OH)<sub>2</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 37.64%; Average Na<sup>+</sup> Removal = 43.39%; Average Observation Period = 24 h);
  - i. Category C2a: n-Mg(OH)<sub>2</sub>@Ca(OH)<sub>2</sub>@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 46.93%; Average Na<sup>+</sup> Removal = 27.92%; Average Observation Period = 24 h);
- c. Category C3: n-Mg(OH)<sub>2</sub>@Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 38.77%; Average Na<sup>+</sup> Removal = 15.04%; Average Observation Period = 24 h);
- d. Category C4: n-Mg(OH)<sub>2</sub>@K<sub>2</sub>CO<sub>3</sub>@Al<sub>2</sub>O<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 47.54%; Average Na<sup>+</sup> Removal = 39.47%; Average Observation Period = 24 h);
  - i. Category C4a: n-Mg(OH)<sub>2</sub>@K<sub>2</sub>CO<sub>3</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 25.10%; Average Na<sup>+</sup> Removal = 9.15%; Average Observation Period = 24 h);
  - ii. Category C4b: n-Mg(OH)<sub>2</sub>@K<sub>2</sub>CO<sub>3</sub>@Al(OH)<sub>3</sub>@MnO<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 29.24%; Average Na<sup>+</sup> Removal = 7.37%; Average Observation Period = 24 h);
- e. Category C5: n-Mg(OH)<sub>2</sub>@MnO<sub>2</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 29.86%; Average Na<sup>+</sup> Removal = 7.26%; Average Observation Period = 24 h);

### **SD4 Category D: n-Zn(OH)<sub>2</sub> polymers**

- a. Category D1: n-Zn(OH)<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 3.73%; Average Na<sup>+</sup> Removal = 3.69%; Average Observation Period = 24 h);
  - i. Category D1a: n-Zn(OH)<sub>2</sub>@Al(OH)<sub>3</sub> polymer; (Average Cl<sup>-</sup> Removal = 23.22%; Average Na<sup>+</sup> Removal = 13.97%; Average Observation Period = 60 h);
  - ii. Category D1b: n-Zn(OH)<sub>2</sub>@Al(OH)<sub>3</sub>@MnO<sub>2</sub> polymer; (Average Cl<sup>-</sup> Removal = 28.89%; Average Na<sup>+</sup> Removal = 4.01%; Average Observation Period = 24 h);

## SD5 Category Assessment

### SD5.1 Category A

The initial ZVI desalination, was observed using Fe(a,b,c) polymers, where the desalination was measured as Cl<sup>-</sup> ion removal (Na<sup>+</sup> ion removal was not measured). Later studies established EC (electrical conductivity) decline, and both Na<sup>+</sup> and Cl<sup>-</sup> ion removal. Category A1, Fe(a,b,c) polymers show very high levels, and rates, of Cl<sup>-</sup> removal, combined with a high selectivity for Cl<sup>-</sup> ion removal, relative to Na<sup>+</sup> removal.

Combining the Fe(a,b,c) polymer with urea (Category A2), reduces the overall Cl<sup>-</sup> ion removal rate, while increasing the Na<sup>+</sup> ion removal rate, to provide a slight selectivity in favor of Cl<sup>-</sup> removal. Addition of CaO (Category A2a), or K<sub>2</sub>CO<sub>3</sub> (Category A2b), reduced the Cl<sup>-</sup> ion removal rate, while increasing selectivity in favor of Cl<sup>-</sup> ion removal.

Combining the Fe(a,b,c) polymer with tartaric acid (Category A3), increases the Cl<sup>-</sup> ion removal rate, while removing no Na<sup>+</sup> ions. Addition of CaO + another organic acid, resulted in high levels of both Cl<sup>-</sup> and Na<sup>+</sup> ion removal (Categories A3a to A3c). Addition of ZnO, was found to maintain high levels of Cl<sup>-</sup> ion removal, while creating a preferential Na<sup>+</sup> ion removal.

Combining the Fe(a,b,c) polymer with malic acid (Category A4), removed Cl<sup>-</sup> ions, while removing no Na<sup>+</sup> ions. The rate of Cl<sup>-</sup> ion removal was significantly slower than when tartaric acid was present. Addition of CaO resulted in increases in Cl<sup>-</sup> ion removal combined with Na<sup>+</sup> removal (Category A4a,A4b). Addition of MnO<sub>2</sub> + HCOOH was found to both increase Cl<sup>-</sup> ion removal and Na<sup>+</sup> removal, while creating a preferential selectivity for Na<sup>+</sup> ion removal (Category A4c).

Combining the Fe(a,b,c) polymer with citric acid (Category A5), removed Cl<sup>-</sup> ions, while removing minimal Na<sup>+</sup> ions. Addition of CaO + ZnO created a preferential selectivity for Na<sup>+</sup> removal (Category 5a). Addition of CaO + MnO<sub>2</sub> + HCOOH was found to both increase Cl<sup>-</sup> ion removal and Na<sup>+</sup> removal, while creating a slight preferential selectivity for Na<sup>+</sup> ion removal (Category A5c). Addition of CaO + K<sub>2</sub>CO<sub>3</sub> resulted in high levels of Cl<sup>-</sup> ion removal, with preferential Cl<sup>-</sup> removal (Category A5b).

Combining the Fe(a,b,c) polymer with ZnO (Category A6), preferentially removed Na<sup>+</sup> ions. Addition of CaO resulted in Cl<sup>-</sup> removal without Na<sup>+</sup> removal (Category A6a, A6e). Addition of Al<sup>0</sup> reversed this change resulting in preferential Na<sup>+</sup> removal (Category A6b).

Combining the Fe(a,b,c) polymer with CaO + gallic acid (Category A7a, A7b), results in high levels of Na<sup>+</sup> and Cl<sup>-</sup> ion removal. Addition of K<sub>2</sub>CO<sub>3</sub> + MgCO<sub>3</sub> increased the selectivity for Cl<sup>-</sup> removal (Category A7c). Combining the Fe(a,b,c) polymer with CaO + MnO<sub>2</sub> + HCOOH resulted in high levels of Cl<sup>-</sup> + Na<sup>+</sup> ion removal with preferential Na<sup>+</sup> removal (Category A7d). Addition of MgCO<sub>3</sub>, increased Cl<sup>-</sup> removal, while reducing Na<sup>+</sup> removal (Category A7e). Replacing MgCO<sub>3</sub> with K<sub>2</sub>CO<sub>3</sub>, would not be expected to increase the available number of sites for Cl<sup>-</sup> removal, but would be expected to decrease Na<sup>+</sup> removal, as K<sup>+</sup> ions compete with Na<sup>+</sup> ions. This is the observed position (Category A7f).

Increasing the availability of K<sub>2</sub>CO<sub>3</sub>, MgCO<sub>3</sub>, and CaCO<sub>3</sub> in the sol-gel formulation was found to maintain a high level of Na<sup>+</sup> ion removal, while substantially reducing the ability of the polymer to remove Cl<sup>-</sup> ions (Category A7g). Elsewhere it has been established that OH<sup>-</sup> ions in metal polymers, are preferentially replaced by HCO<sub>3</sub><sup>-</sup> and CO<sub>3</sub><sup>2-</sup>, when the concentration of HCO<sub>3</sub><sup>-</sup> in the water exceeds a critical level [155]. Replacement of OH<sup>-</sup> with CO<sub>3</sub><sup>2-</sup> would create an additional negative site, which could be used for Na<sup>+</sup> ion removal. This model may provide an explanation for the ion removal selectivity in Category A7g).

Category A8, combines FeSO<sub>4</sub> with MgSO<sub>4</sub> and produces a polymer with a high Na<sup>+</sup> ion removal potential, and a low Cl<sup>-</sup> ion removal potential. Adding ZnO, Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> and CaO, increases the removal of Cl<sup>-</sup> ions, while removing no Na<sup>+</sup> ions (Category A8a). Removing the CaO ingredient, allows some removal of both Na<sup>+</sup> and Cl<sup>-</sup> ions (Category A8b).

Addition of Al(OH)<sub>3</sub> to FeSO<sub>4</sub> + MgSO<sub>4</sub>, retained a selectivity in favor of Na<sup>+</sup> ion removal, while substantially reducing the rate of Na<sup>+</sup> ion removal (Category A8c). Addition of K<sub>2</sub>CO<sub>3</sub> resulted in equal removal of Cl<sup>-</sup> and Na<sup>+</sup> ions while increasing the rate of Na<sup>+</sup> ion removal (Category A8d). Replacement of K<sub>2</sub>CO<sub>3</sub> with CaO + CaCO<sub>3</sub> + Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>, resulted in a substantially increased Cl<sup>-</sup> ion removal coupled with a decreased Na<sup>+</sup> removal (Category A8e). This observation is consistent with H<sup>+</sup> being replaced with Mg<sup>2+</sup> or Al<sup>3+</sup>, preferentially relative to Na<sup>+</sup>. This has the effect of increasing the availability of sites for Cl<sup>-</sup> ion removal while decreasing the availability of sites for Cl<sup>-</sup> ion removal.

Removing the  $\text{CaCO}_3 + \text{Al}_2(\text{SO}_4)_3$  has the effect of reducing the  $\text{Cl}^-$  ion removal while increasing the amount of  $\text{Na}^+$  removed (Category A8f). Removing the  $\text{Al(OH)}_3$  has the effect of increasing  $\text{Cl}^-$  removal, while substantially reducing  $\text{Na}^+$  removal (Category A8g). This is interpreted as indicating that  $\text{Na}^+$  removal is a function of the availability of  $-\text{O}^-$  sites. Adding both  $\text{CaCO}_3$  and K-feldspar has the effect of both increasing  $\text{Cl}^-$  removal and  $\text{Na}^+$  removal (Category A8h). The later increase may result from the replacement of  $-\text{OH}$  with  $-\text{CO}_3^-$ . Replacing K-Feldspar with  $\text{MgCO}_3 + \text{ZnO} + \text{Al}_2(\text{SO}_4)_3 + \text{Al(OH)}_3$  has the effect of increasing  $\text{Na}^+$  removal while decreasing  $\text{Cl}^-$  removal (Category A8i).

These observations indicate that there is no simple solution to the determination of preferred selectivity, coupled with a high rate constant. Selectivity appears to relate directly to the availability of  $-\text{OH}$  sites (Figure A1) and their replacement by one or more of  $-\text{CO}_3^-$ ,  $-\text{O}-\text{Mg}^+$ , or  $-\text{O}-\text{Ca}^+$ , or  $-\text{O}-\text{Zn}^+$ , or  $-\text{O}-\text{Al}^{2+}$ .

## SD5.2 Category B

$\text{CaO}$  based polymers (Category B) demonstrate preferential removal of  $\text{Cl}^-$  ions relative to  $\text{Na}^+$  ions. This selectivity increases in the presence of Al ions,  $\text{K}_2\text{CO}_3$  and  $\text{MnO}_2$  (Category B2, B3a).  $\text{ZnO}$  appear to leave the intrinsic selectivity unchanged (Category B3), though addition of  $\text{K}_2\text{CO}_3$  in the presence of  $\text{ZnO}$ , reduces ion removal, and increases the selectivity for  $\text{Cl}^-$  ion removal (Category B3a).

## SD5.3 Category C

Category C1 and C3 confirmed that a combination of  $\text{MgSO}_4 + \text{Al}_2\text{O}_3$ , removed both  $\text{Cl}^-$  and  $\text{Na}^+$  ions with a preferential selectivity for  $\text{Cl}^-$  ion removal. Addition of  $\text{CaO}$  increased  $\text{Na}^+$  ion removal (Category C2). Increasing the proportion of Al in the formulation increased the total amount of  $\text{Cl}^-$  removed, while reducing the amount of  $\text{Na}^+$  removed (Category C2a). Addition of  $\text{MnO}_2$  or  $\text{K}_2\text{CO}_3$ , did not alter the amount of  $\text{Cl}^-$  removed, but decreased the proportion of  $\text{Na}^+$  removed (Category 4, C5).

## SD5.4 Category D

Category D1 demonstrates that  $\text{ZnO}$  ( $\text{Zn}(\text{OH})_2$ ), when present on its own is an ineffective desalination agent, which removes  $\text{Cl}^-$  and  $\text{Na}^+$  ions in approximately equal proportions. Adding  $\text{Al(OH)}_3$  to  $\text{ZnO}$ , increases both  $\text{Na}^+$  and  $\text{Cl}^-$  removal, creating a preferential selectivity for  $\text{Cl}^-$  ion removal (Category D1a). Modifying this formulation by adding  $\text{MnO}_2$  (Category D2), increases both the amount of  $\text{Cl}^-$  removed and the selectivity for  $\text{Cl}^-$  removal.

## Section SE Microscopic Analysis of Metal Polymers

### SE1 Trials F39, F74 and F78

Fluids associated with Trial F39 were not examined microscopically. No polymer spheres were observed in the product water, or evaporated product water from Trial F74. Instead, two distinct crystalline minerals dominated the evaporated product water (Supplementary Information, Figure S16). Both minerals were based on  $\text{Mg}(\text{OH})_2$  metal polymers. They were a dumbbell crystal structure ( $(\text{Mg}(\text{OH})_2)_n \text{NaOH} (\text{H}_2\text{O})_m$ ) and acicular crystal flowers ( $2\text{Mg}(\text{OH})_2\text{MgCl}_2 (\text{H}_2\text{O})_2$ ) (Supplementary Information, Figure S16a, S16b). Examination of the product water established the presence of settled (formerly entrained) dark agglomerates constructed from the aggregation of smaller spheres, and smaller entrained spheres (Supplementary Information, Figure S16c, S16d). It was not possible to determine, if the spheres were hollow and gas filled. The absence of a strong refractive difference between the center and outside margin of the spheres, indicates that either the spheres are solid or that their interior fluid is dominantly water. Three sizes of spheres were observed (Figure S17) where the smallest sized spheres aggregated to form the medium sized spheres. The largest sized spheres were formed from the aggregation of the medium sized spheres.

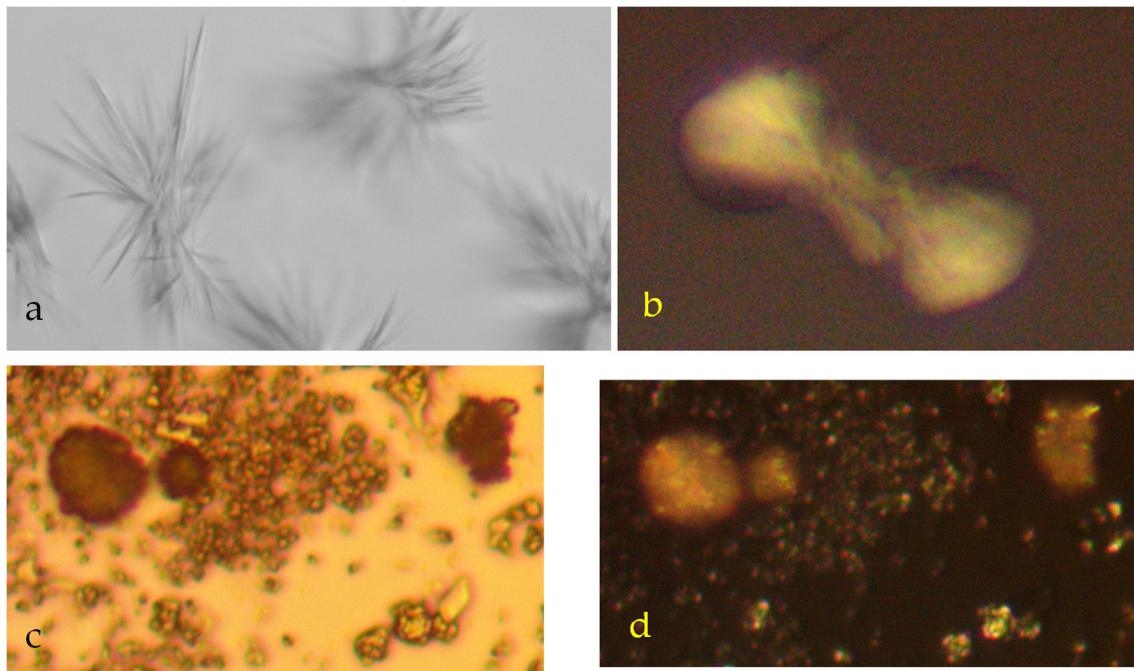


Figure S16. Trial F74 precipitates. (a), Acicular polymer flowers of  $(2\text{Mg}(\text{OH})_2\text{MgCl}_2(\text{H}_2\text{O})_2)$ , or  $\text{Mg}_2(\text{OH})_3\text{Cl}(\text{H}_2\text{O})_4$ ; Transmitted light; Field of view = 123 microns; (b), Dumbbell polymer crystals of  $((\text{Mg}(\text{OH})_2)_n \text{NaOH}(\text{H}_2\text{O})_m)$ ; Cross polarized light; Field of view = 43.5 microns; (c), Entrained polymer precipitates; Dark precipitates are  $(\text{Fe}(\text{OH})_n \text{Mg}(\text{OH})_2 \text{NaOH})$  composite polymer aggregates, light precipitates are  $(\text{Mg}(\text{OH})_2\text{NaOH})$  polymers; The larger transparent platy crystals are  $\text{MgSO}_4$ . transmitted light; Field of view = 86.6 microns; ; (d), Entrained precipitates; Cross polarized light light; Field of view = 86.6 microns.

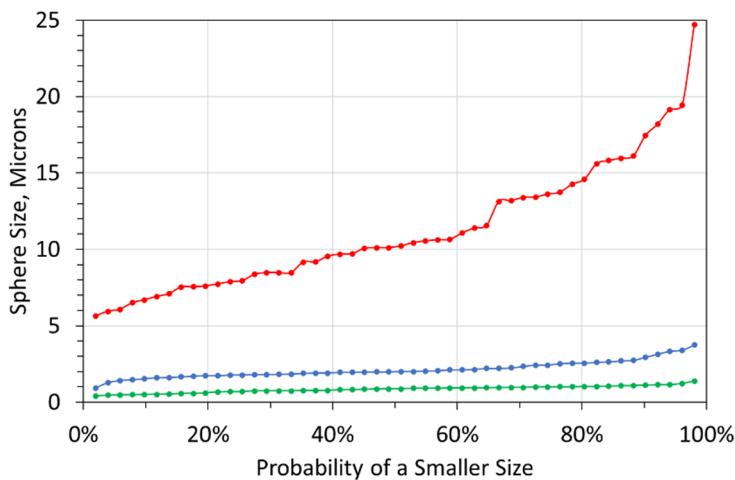


Figure S17. F74. Size distribution of the 3 distinct groups of polymer spheres. Green = smallest size (mean = 0.84 microns; standard deviation = 0.22); Blue = intermediate size (mean = 2.12 microns; standard deviation = 0.55); Red = largest size (mean = 11.21 microns; standard deviation = 4.11); Number of measurements in each category = 50.

The dumbbell and acicular crystal morphologies (Supplementary Information, Figure S16a, S16b) were not observed in Trial F78. Instead, the settled FeOOH precipitate was amorphous. However, the

water contained entrained, colorless, translucent, anisotropic, biaxial, monoclinic, crystals (Supplementary Information, Figure S18), around 8-10 microns in length. If all the  $Mg^{2+}$  was precipitated, then to account for the removed  $Na^+$  ions, the molar ratio is 2.22Na removed to 1Mg.. These crystals are interpreted as a sodium magnesium sulphate (Blodite, bloedite), e.g.,  $Na_{2.2}Mg(SO_4)_{2.1}(H_2O)_z$ . This structuring occurs in the presence of an excess of sulphate ions. It allows for higher levels of  $Na^+$  removal as blodite transforms to vanthoffite ( $Na_6Mg(SO_4)_4$ ) [156]. The general Blodite structure takes the form  $Na_2M(SO_4)_2(H_2O)_z$ , where M is one or more of Mg, Zn, Ni, Co, Fe and Mn [157,158,159].

In this example, the polymer ingredients created two overlapping quaternary systems. System 1 =  $NaCl + MgCl_2 + Na_2SO_4 + MgSO_4 + H_2O$ ; System 2 =  $NaCl + FeCl_2 + Na_2SO_4 + FeSO_4 + H_2O$ , containing divalent metals. The resulting potential structures are  $[(Na_2SO_4)_n (MSO_4)_m (H_2O)_y]$ . The presence of  $Fe^{3+}$  creates a third overlapping system; System 3 =  $NaCl + FeCl_3 + Na_2SO_4 + Fe(OH)_{3-y}(SO_4)_y + (Fe^{2+})_a (Fe^{3+})_b (OH)_{a+b-y}(SO_4)_y + Fe_{1-y}(OOH)(SO_4)_y + H_2O$ . In F78 the third system only produced amorphous iron polymers.

The microscopic observations for Trials F74 and F78 (Category A8), suggest that in the presence of Mg and  $SO_4$ , selective  $Na^+$  removal, is a result of incorporation in  $Mg SO_4$  crystallite structures.

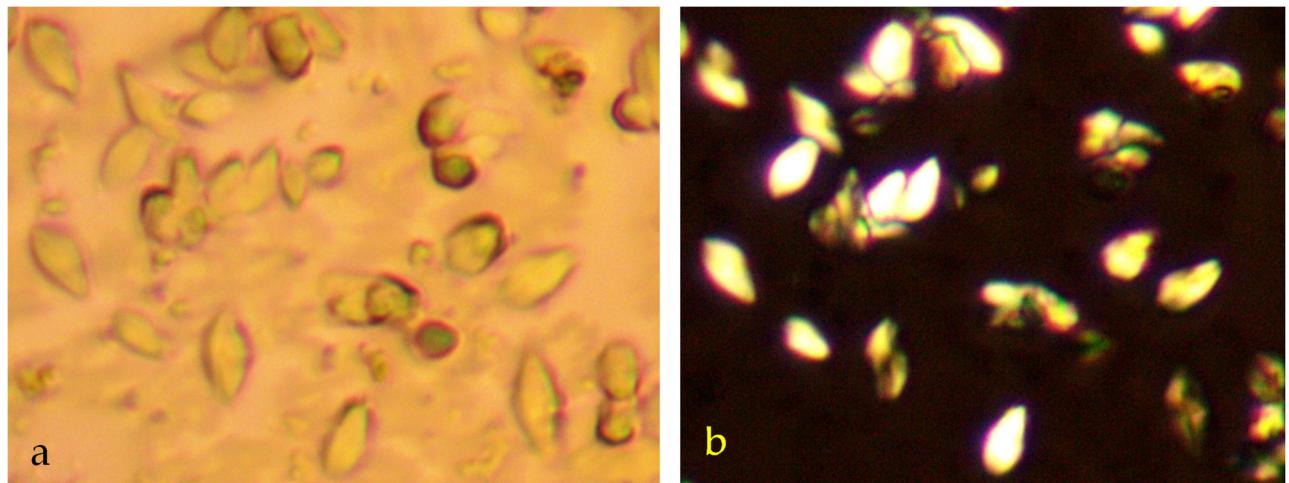


Figure S18. Trial F78. Entrained crystals. (a), transmitted light. Field of view = 68 microns; (b), crossed polarised light. Field of view = 67 microns.

### SE2 Selectivity: $Cl^-$ Ion Removal no $Na^+$ Ion Removal

Trials F5, F8, F11, F25, F29, F33, F52, F68, and F87 demonstrated  $Cl^-$  ion removal with no effective  $Na^+$  ion removal.

#### SE2.1 Trial F5 (Category A3)

This trial, contained a polymer constructed from tartaric acid and  $FeSO_4$ . It reduced the  $Cl^-$  ion content from  $22.11 \text{ g L}^{-1}$  to  $1.51 \text{ g L}^{-1}$ , over a period of 0.1 h. This product water was not retained for microscopic analysis.

#### SE2.2 Trial F8 (Category A4)

This trial, contained a polymer constructed from malic acid and  $FeSO_4$ . It reduced the  $Cl^-$  ion content from  $22.11 \text{ g L}^{-1}$  to  $11.67 \text{ g L}^{-1}$ , over a period of 0.1 h. This product water was not retained for microscopic analysis.

### SE2.3 Trial F11 (Category A5)

This trial, contained a polymer constructed from citric acid and FeSO<sub>4</sub>. It reduced the Cl<sup>-</sup> ion content from 22.11 g L<sup>-1</sup> to 1.33 g L<sup>-1</sup>, over a period of 0.1 h. Evaporation of the product water established the presence of hexagonal platy crystals (Supplementary Information, Figure S19a; S19b), and small entrained spheres (Supplementary Information, Figure S19b) within the water. Quantities of amorphous FeOOH and NaCl were observed. The NaCl resulted from the release of fluids from spheres of FeOOH, which disintegrated during evaporation.

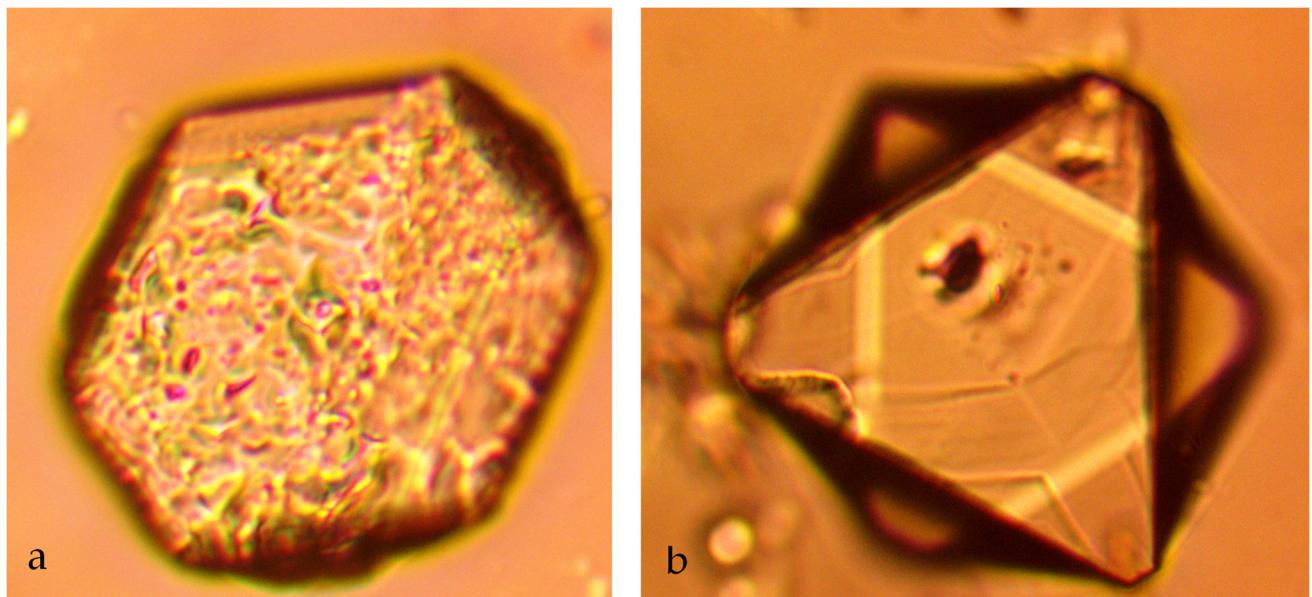


Figure S19. Trial F11: Evaporated water crystals (interpreted as a form of sodium citrate), indicating that the product water was depleted in Cl<sup>-</sup> ions. (a), hexagonal crystal. Field of view = 80 microns; (b), twinned hexagonal crystals, and small spheres in the surrounding water. Field of view = 130 microns

### SE2.4 Trial F25 (Category A4a)

This trial, contained a polymer constructed from malic acid, CaO, ZnO and FeSO<sub>4</sub>. It resulted in 71% Cl<sup>-</sup> removal, with no effective Na<sup>+</sup> removal, from a feed water containing 2.76 g Cl<sup>-</sup> L<sup>-1</sup> and 1.23 g Na<sup>+</sup> L<sup>-1</sup>. The product water contained 0.8 g Cl<sup>-</sup> L<sup>-1</sup> and 1.23 g Na<sup>+</sup> L<sup>-1</sup> after 24 h. A number of observations were made. After about 2 h, the water contained a high concentration of anisotropic, spherical crystals (Figure S20a). After 5 h, the concentration of entrained particles, within the water, had reduced substantially, and many of the crystals were elongate (Figure S20b). This change, was accompanied by, a major increase in the size of the entrained particles (Figure S20c). The larger entrained particle distribution was multi-modal (Figure S20c). After 2 h the settled precipitate consisted of aggregated particles (Figure S20d, S20e), which were smaller than the entrained particles (Figure S20c). After 5 h the settled precipitate consisted of aggregated particles (Figure S20d, S20f), which were smaller than the entrained particles (Figure S20c). The settled particles, also included, a number of larger, hollow, spherical particles (Figure S20d; S20f). These larger particles are anisotropic (Figure S20b).

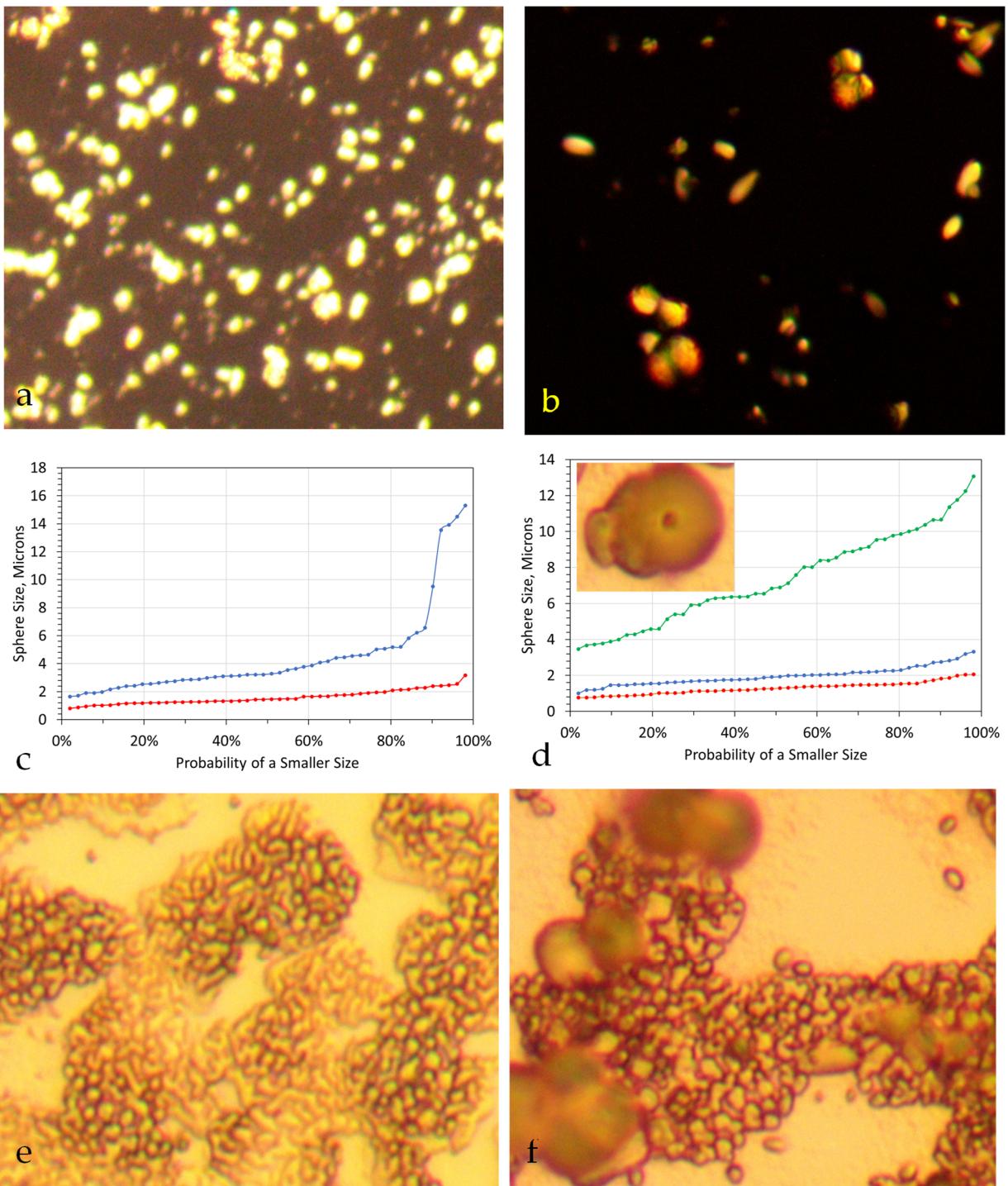


Figure S20. Trial F25. Entrained particles. Cross polarized light; (a), after 2 h. Field of view = 55 microns; (b), after 5 h. Field of view = 87 microns; (c), size distribution of entrained particles; Red = after 2 h (mean = 1.58 microns; standard deviation = 0.50); Blue = after 5 h (mean = 4.47 microns; standard deviation = 3.72); (d), size distribution of settled particles; Aggregated particles: Red = after 2 h (mean = 1.29 microns; standard deviation = 0.34); Aggregated particles: Blue = after 5 h (mean = 1.97 microns; standard deviation = 0.51); Isolate particles: Green = after 5 h (mean = 7.36 microns; standard deviation = 2.56). Inset is of a larger, isolate, settled, hollow, spherical particle (green distribution) after 5 h (Field of view = 20 microns; particle diameter 13.4 microns); (e) Settled precipitate after 2 h. Field of view = 52 microns; (f) Settled precipitate after 2 h. Field of view = 57 microns;

### SE2.5 Trial F29 (Category A7)

This trial, contained a polymer constructed from CaO and FeSO<sub>4</sub>. It reduced the Cl<sup>-</sup> ion content from 12.14 g L<sup>-1</sup> to 6.5 g L<sup>-1</sup>, over a period of 24 h, with no removal of Na<sup>+</sup> ions. This product water was not retained for microscopic analysis.

### SE2.6 Trial F33 (Category A2a)

This trial, contained a polymer constructed from CaO, Urea and FeSO<sub>4</sub>. It reduced the Cl<sup>-</sup> ion content from 30.14 g L<sup>-1</sup> to 12.46 g L<sup>-1</sup>, over a period of 24 h, with no removal of Na<sup>+</sup> ions. This product water was not retained for microscopic analysis.

### SE2.7 Trial F52 (Category A8g)

This trial, contained a polymer constructed from CaO, MgSO<sub>4</sub> and FeSO<sub>4</sub>. It reduced the Cl<sup>-</sup> ion content from 30.14 g L<sup>-1</sup> to 12.46 g L<sup>-1</sup>, over a period of 24 h, with no removal of Na<sup>+</sup> ions. This product water was retained for microscopic analysis. A major difference was observed between the evaporated product of the feed and product waters (Figure S21). The feed water produced a highly saline evaporated product (Figure S21a). The evaporated product water was less saline and included anisotropic, spherical and hexagonal crystals (Figure S21b).

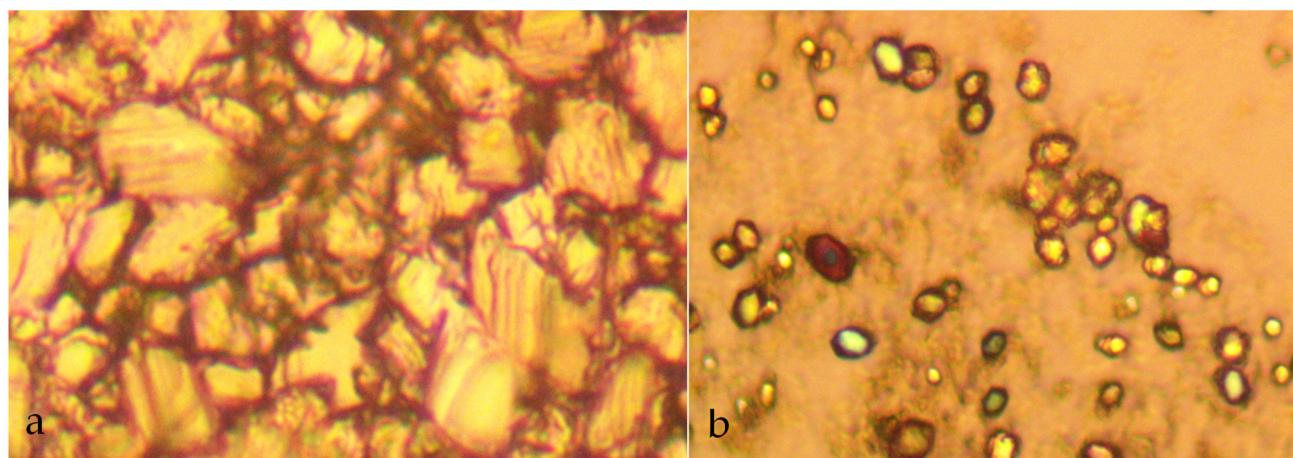


Figure SE21. Trial F52: (a), Evaporated feed water. Field of view = 73 microns; (b), Evaporated product water. Field of view = 120 microns.

### SE2.8 Trial F68 (Category C3)

This trial, contained a polymer constructed from MgSO<sub>4</sub> and Al(OH)<sub>3</sub>. It reduced the Cl<sup>-</sup> ion content from 21.87 g L<sup>-1</sup> to 9.33 g L<sup>-1</sup>, over a period of 24 h, with no removal of Na<sup>+</sup> ions. The product water contained entrained anisotropic particles (Supplementary Information, Figure S22). These particles settled to form short chains of spherical polymer particles (Figure S23a), which coalesced to form larger chains (Figure S23b).

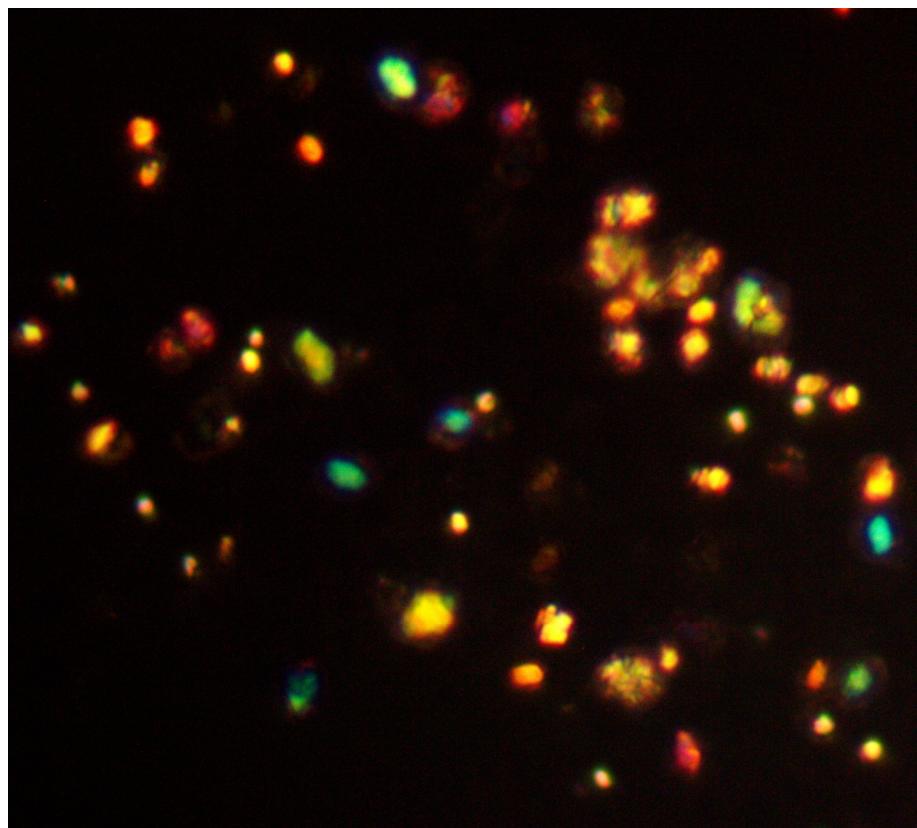


Figure S22. Trial F68: Entrained anisotropic Mg-Al polymers in the product water. Field of view = 126 microns

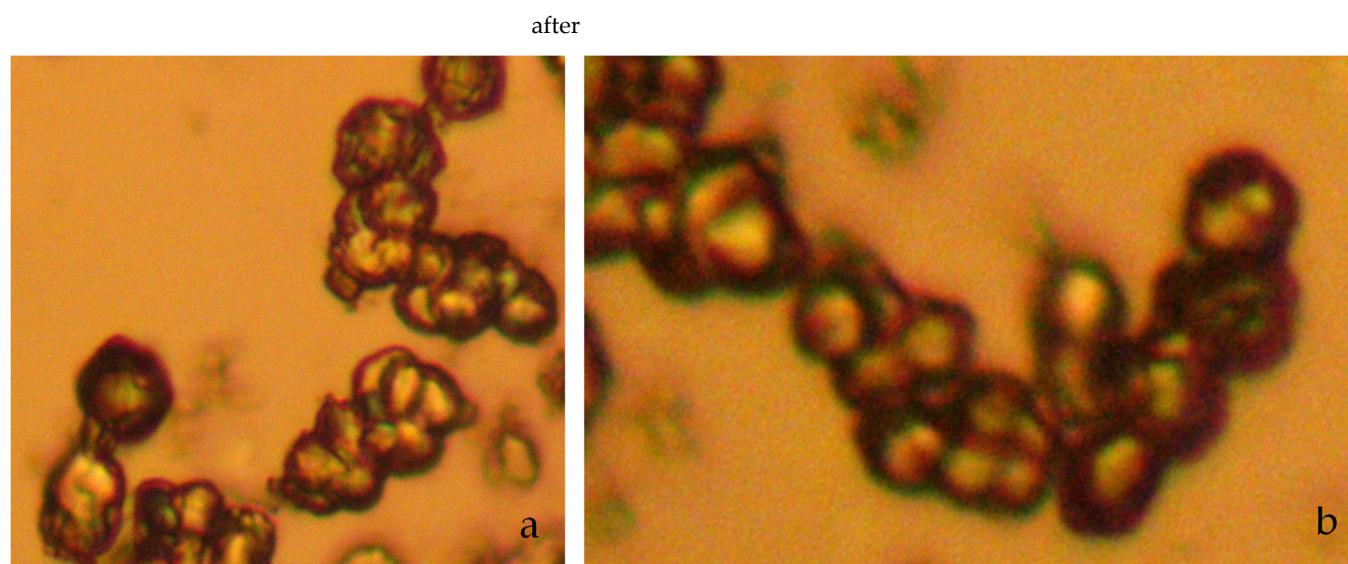


Figure S23. F68: Mg-Al polymer chains constructed from polymer spheres. (a), isolate incipient shorter chains. Field of view = 58 microns; (b), Longer chain formed from the aggregation of shorter chains. Field of view = 37 microns.

### SE2.9 Trial F87 (Category A8a)

This trial, contained a polymer constructed from CaO, ZnO, Al(OH)<sub>3</sub> and FeSO<sub>4</sub>. It reduced the Cl<sup>-</sup> ion content from 7.96 g L<sup>-1</sup> to 5.39 g L<sup>-1</sup>, over a period of 24 h, with no removal of Na<sup>+</sup> ions. This product water was not retained for microscopic analysis.

### SE3 Trial F1, F3, F7, F10

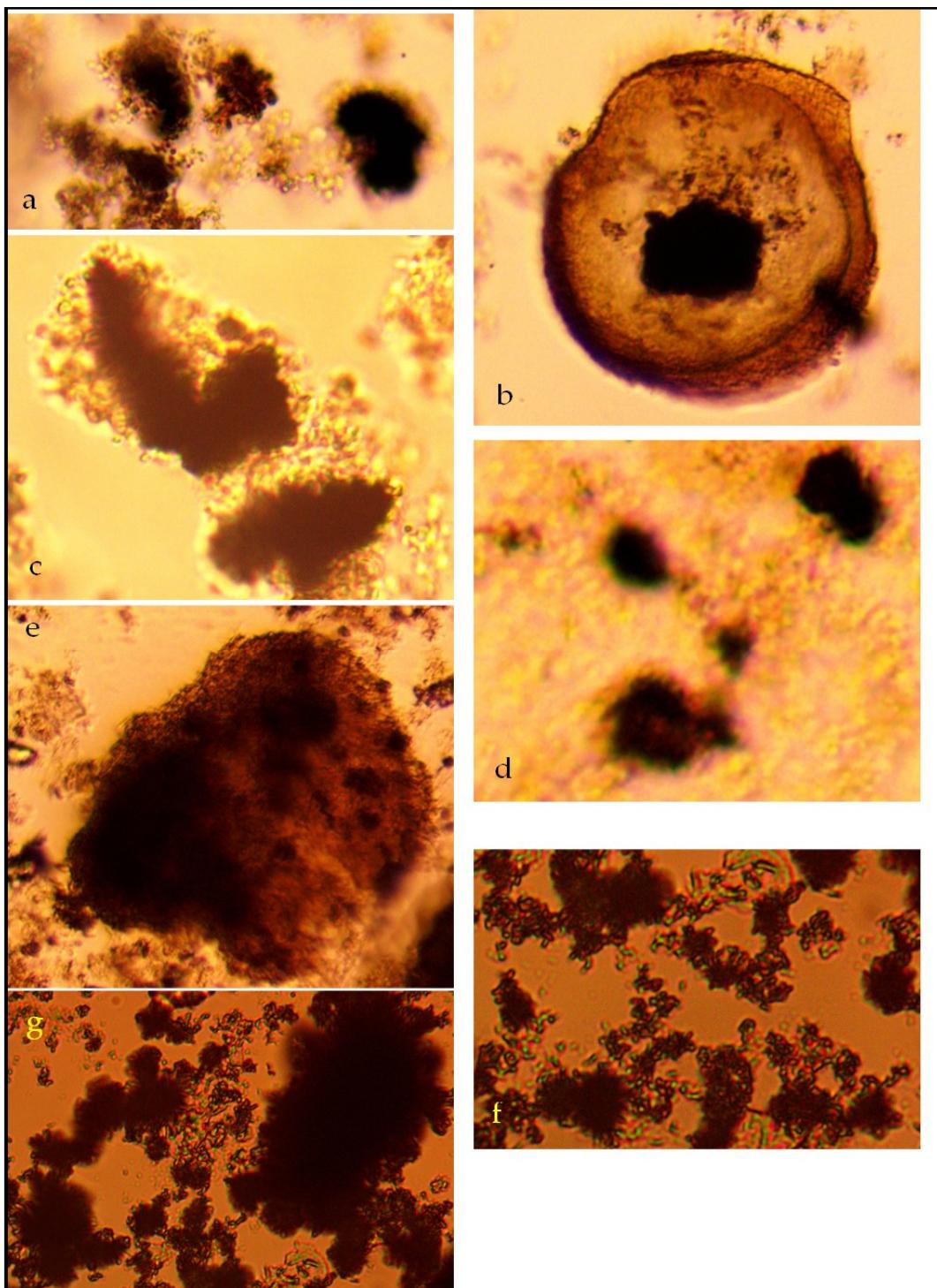


Figure S24. Fe(a,b,c) sphere relationships with MnO<sub>2</sub>. (a), Trial F1, Fe(a,b,c) spheres coalescing around MnO<sub>2</sub> particles. Field of view = 53 microns; (b), Trial F1, Colloidal spherical particle formed from Fe(a,b,c) spheres coalescing around MnO<sub>2</sub> particle. Field of view = 50 microns; (c), Trial F3, Fe(a,b,c) spheres coalescing around MnO<sub>2</sub> particles. Field of view = 107 microns; (d), Trial F7, Fe(a,b,c) spheres coalescing around MnO<sub>2</sub> particles. Field of view = 24.7 microns; (e), Trial F7, Fe(a,b,c) spheres coalescing around MnO<sub>2</sub> particles, to form a single colloidal particle. Field of view = 63 microns; (f), Trial F10, Fe(a,b,c) spheres coalescing around MnO<sub>2</sub> particles. Field of view = 87 microns; (g), Trial F10, Fe(a,b,c) spheres coalescing around MnO<sub>2</sub> particles. Field of view = 140 microns;