## **EES Solar**



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# Salt ion triggered hydration chemistry for saltrejecting solar-driven water evaporation

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The escalating global freshwater scarcity necessitates sustainable and efficient desalination technologies, yet conventional methods remain constrained by high energy consumption and operational complexity. Solar-driven water evaporation has emerged as a promising alternative, but its practical deployment is hindered by salt fouling, which restricts the evaporation efficiency and durability under high-salinity conditions. This review highlights salt ion triggered hydration chemistry as pivotal for hydrogel-based evaporators, with a focus on two strategies: Hofmeister effect driven hydration and anti-polyelectrolyte effect enabled hydration. These two approaches have demonstrated remarkable potential in overcoming the limitations of conventional desalination methods, such as high energy consumption and salt fouling through tailoring water transport pathways and enhancing salt tolerance. The key mechanism for salt rejection is that these polymers can bind with salt ions to suppress salt precipitation, release more weakly bound water to reduce the evaporation enthalpy and improve the brine pumping ability of evaporators in saline water. Hydration chemistry has thus emerged as a cornerstone for the design of next-generation hydrogel evaporators, advancing water purification, which aligns with global sustainability goals. We believe this review will bring new understanding into the chemical design of hydrogel based solar-driven evaporation systems.

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### **Broader context**

The escalating global freshwater crisis underscores the urgent need for sustainable and efficient desalination technologies. Conventional methods, however, are often hindered by high energy consumption and operational complexity. Solar-driven interfacial water evaporation has emerged as a promising alternative, offering a sustainable, low-cost, and eco-friendly route to mitigate freshwater scarcity. By localizing solar heat at the evaporator surface, this approach efficiently produces freshwater using only solar energy and seawater. Nevertheless, a critical challenge persists: while high evaporation rates are essential in producing freshwater, they often accelerate salt precipitation, which in turn undermines efficiency by obstructing water channels and blocking sunlight. This review focuses on salt-ion-regulated hydration chemistry as an emerging strategy to suppress salt crystallization while maintaining high evaporation rates, even in highly saline brines. With tailored polymer designs, salt ions can play a beneficial role in enhancing the hydration of some specific hydrogel evaporators. We classify the underlying mechanisms into two categories: Hofmeister-effect-induced hydration and anti-polyelectrolyte-effect-induced hydration. Each approach is systematically analyzed with a detailed discussion of its chemical principles and operational mechanisms. We anticipate that this review will offer new insights into the chemical design of advanced hydrogel evaporators and propel the development of high-efficiency, salt-rejecting solar desalination technologies.

### 1. Introduction

The global freshwater crisis, exacerbated by population growth, industrialization, and climate change, has rendered water scarcity a critical challenge of the 21st century. While seawater desalination offers a viable solution, conventional technologies such as reverse osmosis and multi-stage flash distillation remain energy-intensive, costly, and infrastructure-dependent, limiting their scalability in resource-constrained regions. In recent years, solar-driven water evaporation technology has emerged as a sustainable alternative, utilizing solar energy to locally heat the water at the air–water interface for efficient vapor generation. Despite their promising potential, evaporators face inherent limitations arising from the salinity

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of water, specifically: (1) prolonged evaporation leads to the progressive increase of salt concentration and ultimately causes the accumulation of salt crystals, which reduce the evaporation efficiency by scattering incident sunlight and clogging water channels; 9,10 (2) the dissolved salt ions bind with water robustly and thus raise the activation energy required for water evaporation. These factors suppress the evaporation performance of traditional evaporators and particularly compromise the long-term evaporation performance of evaporators under hypersaline conditions. 11,12

Substantial research efforts have been dedicated to mitigating salt accumulation through advanced structural engineering, mainly employing the following methods:<sup>13</sup> (1) spatially confining salt crystallization at the edge region of the solar surface by geometric design, which keeps the central zone clean;<sup>14</sup> (2) design of Janus structures with asymmetric

wettability to suppress the salt ion migration toward the solar interface;<sup>15,16</sup> (3) continuous salt dissolution processes through dynamic fluid exchange with surrounding seawater. While these innovations have significantly improved salt tolerance, critical limitations still persist: (1) complicated structures like hierarchical channels, surface morphology or specific geometries might increase the manufacturing complexity and deform their shape during repeated use;<sup>17,18</sup> (2) thermal energy would be lost during the liquid exchange process, compromising the evaporation efficiency.<sup>19,20</sup> Besides, although the accumulation of precipitated salt crystals is mitigated, these methods cannot address the problem of high energy demand for water



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vaporization induced by dissolved salt ions. Consequently, developing straightforward approaches to concurrently address precipitated salt and dissolved salt remains a critical challenge.

One emerging strategy to address the salt issues is through salt ion triggered hydration chemistry, focusing on enhancement of salt tolerance by tailoring the interactions between polymer, salt ions and water based on molecular design.21 In this strategy, hydrogels have emerged as a pivotal candidate due to their unique three-dimensional polymeric networks capable of storing substantial amounts of water and regulating the state of salt and water.22,23 Compared with structural engineering approaches, the hydration chemistry strategy offers a distinct advantage by addressing both issues of precipitated salt crystals and dissolved salt simultaneously.24 These specific polymers can bind with salt ions robustly to suppress salt precipitation, while releasing more loosely bound water molecules (intermediate water, IW) to reduce the water evaporation enthalpy of systems; meanwhile, the salt ions dissociate the inter-chain associations, leading to the swelling of the gel, which boosts the adsorption and transportation of brine.<sup>25-27</sup> Such an intrinsic chemical mechanism enables these hydrogels to maintain high evaporation efficiency even in hypersaline environments, avoiding the typical trade-offs between complexity, energy loss, and durability inherent in structural methods.28 Two main principles have caught researchers' eyes in recent years: (1) the Hofmeister effect existed in diverse polyvinyl alcohol (PVA)-based hydrogels,29 and (2) the anti-polyelectrolyte effects mainly existed in zwitterionic polymers. 30,31 It should be noted that these salt responsive polymers usually do not have photothermal conversion ability themselves. Therefore, additional photothermal fillers like polypyrrole (PPy), MXene and graphene oxide (GO) are required to achieve photothermal conversion for solar-driven water evaporation.32-34

This review systematically explores the interplay between hydration chemistry and solar desalination, with a focus on ion-specific regulation in PVA networks enabled by the Hofmeister effect and the salt induced hydration phenomenon existing in zwitterionic polymers based on the anti-polyelectrolyte effect (Fig. 1). By dissecting the hydration mechanisms, performance metrics (e.g., evaporation rates and evaporation enthalpy) and material innovations, this work critically evaluates both the efficiency and durability of solar-driven water evaporation systems. Finally, we outline the potential directions for the future. By unraveling the molecular principles governing the salt mediated hydration capability, this work aims to inspire the chemical design of next-generation hydrogel evaporators that reconcile high evaporation efficiency, sustainability and salt resistance simultaneously.

# 2. General design of the solar-driven water evaporation system

A typical solar-driven water evaporation system consists of two key components: (1) a water transport matrix for continuous brine supply and vapor escape, (2) a photothermal component for solar energy absorption and heat generation. <sup>35,36</sup> The synergy

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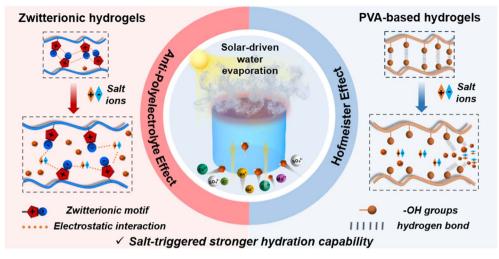


Fig. 1 Schematic illustration of the salt ion triggered hydration chemistry for salt-rejecting solar-driven water evaporation.

of the two components enables a high rate of water evaporation and supply of liquid to the heated region, significantly improving evaporation efficiency.<sup>37</sup>

The efficiency of solar evaporation systems critically depends on the hydrogel's ability to continuously replenish water while expelling accumulated salt ions.<sup>38</sup> Hydrogels, which are three-dimensional polymeric networks saturated with water and possessing the remarkable capacity to regulate the state of salt and water, have emerged as a highly adaptable material platform for solar-driven water evaporation.<sup>39,40</sup> A well-designed water transport network typically features hydrophilic channels to enable capillary-driven water flow and strong hydration capacity to guarantee salt tolerance.<sup>41</sup>

Most hydrogels lack inherent photothermal conversion capabilities.42 To enable efficient solar energy harvesting, photothermal fillers that can convert sunlight into heat through broad-spectrum light absorption are incorporated into hydrogels.42 For example, most photothermal materials, including polymers (e.g., PPy), carbon-based materials (e.g., GO, CNTs) and plasmonic nanoparticles (e.g., Au, Ag) could be added directly into the precursor of gels before polymerization.43 Meanwhile, photothermal polymers like PPy and polyaniline (PANI) can also grow in situ on/in the hydrogel after polymerization.8,44,45 These materials exhibit strong light absorption across the solar spectrum (UV-vis-NIR) due to their intrinsic electronic transitions, surface plasmon resonance or phonon interactions.46 For example, PPy achieves >90% solar absorption *via*  $\pi$ – $\pi$ \* transitions, 47 while MXene's layered structure enhances light trapping through multiple internal reflections.48

# 3. Strategies for chemically activated hydration

The hydration capacity of hydrogels is governed by the chemical structure of polymers. <sup>49</sup> For example, polyvinyl alcohol (PVA) can bind water molecules through hydrogen bonds between hydroxyl groups and water. <sup>50</sup> Zwitterionic polymers (*e.g.*,

polysulfobetaine) exhibit electrostatic interaction induced hydration through their abundant charged moieties (e.g.,  $-N^+$  and  $-SO_3^-$ ), whose hydration ability is even stronger than that through hydrogen bonding.<sup>51</sup> These distinct mechanisms dictate their salt responsiveness: the hydration behavior of PVA in brine is mainly dominated by the Hofmeister effect, while that of zwitterionic polymers exhibits an anti-polyelectrolyte effect in saline water.<sup>52,53</sup>

# 3.1 Hofmeister effect of PVA-based materials for solar-driven water evaporation

**3.1.1 Hofmeister effect.** The Hofmeister effect is originally observed in protein solubility, which has also been widely studied in diverse polymer systems. The correlation of ions and protein solubility depends on the Hofmeister series:  $\operatorname{Cit}^{3-} > \operatorname{SO_4}^{2-} > \operatorname{S_2O_3}^{2-} > \operatorname{H_2PO_4}^{-} > \operatorname{F^-} > \operatorname{CH_3COO^-} > \operatorname{Cl^-} > \operatorname{NO_3}^{-} > \operatorname{Br^-} > \operatorname{I^-} > \operatorname{ClO_4}^{-} > \operatorname{SCN^-}$  and  $\operatorname{NH_4}^+ > \operatorname{K^+} > \operatorname{Na^+} > \operatorname{Li^+} > \operatorname{Mg}^{2+} > \operatorname{Ca}^{2+}$  (Fig. 2). The former ions are well-hydrated kosmotropes that hinder protein solubility. Conversely, the latter ones are poorly hydrated chaotropes that facilitate protein solubility. S

According to the proposed Hofmeister series from protein solubility, typical salt ions can be classified into kosmotropes and chaotropes, critically governing the hydration capability of polymers in saline water.<sup>55</sup> (1) The kosmotropic ions result in the loss of water molecules from polymers, accompanied by the formation of new hydrogen bonds between polymer chains. This process would lead to the aggregation of polymers and a reduction in polymer solubility (called salting out).<sup>56</sup> (2) In contrast, chaotropic ions interact directly with the polymer, resulting in the addition of charges between the polymer chains, thereby promoting its hydration and increasing the solubility of the polymer (called salting in).<sup>57,58</sup> The typical polymers showing an obvious Hofmeister effect include PVA,<sup>59</sup> Chitosan,<sup>60</sup> poly(*N*-isopropylacrylamide) (PNIPAM)<sup>61,62</sup> and so on.<sup>63</sup>

Natural seawater typically contains  $\sim$ 3.5 wt% dissolved salts, dominated by Na $^+$  (10.8 g L $^{-1}$ ), Cl $^-$  (19.3 g L $^{-1}$ ), Mg $^{2+}$  (1.3 g L $^{-1}$ ),

Cit<sup>3-</sup> >  $SO_4^{2-}$  >  $S_2O_3^{2-}$  >  $H_2PO_4^-$  >  $F^-$  >  $CH_3COO^-$  >  $Cl^-$  >  $NO_3^-$  >  $Br^-$  >  $l^-$  >  $ClO_4^-$  >  $SCN^-$  Salting out Salting in  $N(CH_3)_4^+$  >  $NH_4^+$  >  $Cs^+$  >  $Rb^+$  >  $K^+$  >  $Na^+$  >  $Li^+$  >  $Ca^{2+}$  >  $Mg^{2+}$ 

Fig. 2 General Hofmeister series of anions and cations.

and  ${\rm Ca^{2^+}}$  (0.4 g  ${\rm L^{-1}}$ ). <sup>64</sup> These ions will directly influence hydrogel hydration based on the Hofmeister effect on polymer materials. <sup>65</sup> Specifically, the chaotropic ions in brine (Cl $^-$ , Na $^+$ , Mg $^{2^+}$ , Ca $^{2^+}$ ) may induce the salting-in process of evaporators and boost the hydratability, which are key to activating water transport and decreasing the evaporation enthalpy. <sup>28</sup> Conversely, kosmotropic ions (*e.g.*, Cit $^{3^-}$ ) induce hierarchical porosity, which can also optimize capillary-driven water transport. <sup>66,67</sup>

**3.1.2 PVA-based materials for solar-driven water evaporation.** PVA is a typical polymer showing an obvious Hofmeister effect, which has been widely used in solar-driven water evaporation. The Chaotropic ions in seawater (*e.g.*, Cl<sup>-</sup>, Na<sup>+</sup>, Mg<sup>2+</sup> and Ca<sup>2+</sup>) can disrupt the hydrogen bonds between PVA chains, liberating hydroxyl groups to form more IW. The IW is more active than free water (FW) and requires lower energy for evaporation. This ion-mediated hydration simultaneously lowers the evaporation enthalpy and enhances brine uptake, enabling hydrogels to achieve high evaporation rates in high-salinity environments. Besides, the phenomenon will be more significant as the ion concentration increases, showing great potential in the treatment of industrial wastewater or highly concentrated saline water.

For example, the Qiu group designed a highly hydratable hydrogel (PMH) consisting of modified needle coke as a solar absorber and PVA as a hydratable matrix, demonstrating exceptional solar-driven evaporation performance in hypersaline environments.28 In this work, the chaotropic Cl ions in brine are proved to disrupt the hydrogen-bonding networks within PVA chains and liberate more hydroxyl groups of PVA chains in PMH (Fig. 3a).28 Such improved ion-mediated hydration activation of PVA within PMH based on the Hofmeister effect induced two critical results: (1) an elevated intermediate water/free water (IW/FW) ratio effectively lowered the enthalpy requirement for water evaporation (Fig. 3b-d); (2) reduction in PVA crystallinity coupled with enhanced surface hydrophilicity (Fig. 3e).28 These synergistic effects enabled PMH to achieve an exceptional evaporation rate of 3.18 kg m<sup>-2</sup> h<sup>-1</sup> under 1 sun in simulated seawater, surpassing pure water performance by 26%.28 Moreover, the PMH maintained stable evaporation performance (2.9 kg m<sup>-2</sup> h<sup>-1</sup>) during 21 hours of continuous operation, with rapid salt dissolution kinetics effectively preventing crystalline deposition (Fig. 3f).28 This further showed the salt-resistant durability of PMH, which stemmed from salt ion-triggered hydration activation of PVA.28

Li *et al.* developed a PVA/GO hydrogel-based solar steam generator (named as PGD), whose polymer network was further regulated by simply adding chaotropic I<sup>-</sup> ions (named as PGI).<sup>32</sup>

The introduction of the I<sup>-</sup> ions disrupted the hydrogen-bonding networks between PVA chains and increased the number of exposed –OH moieties (Fig. 4a), resulting in the increased content of IW (Fig. 4b) and thus reduction of the water evaporation enthalpy. Molecular dynamics simulation results showed that there were more hydrogen bonds in the PVA system than in the PVA-KI system, which successfully proved the increased number of exposed –OH moieties in PGI due to the Hofmeister effect (Fig. 4c).<sup>32</sup> As a result, the evaporation performance of PGI was improved to 140% of that of PGD, whose evaporation rate reached 2.32 kg m<sup>-2</sup> h<sup>-1</sup> in seawater.<sup>32</sup>

It is worth noting that the Hofmeister effect can not only be used to regulate the hydration ability of hydrogels, but also to regulate their hierarchical pores with the assistance of kosmotropic ions. 66,67,76 As a result of optimized porosity, brine pumping capability might be promoted. For example, Jiang et al. introduced the kosmotropic citrate ions (Cit<sup>3-</sup>) into PVA gels to obtain a sponge-like PVA/graphite composite hydrogel (PGCNG) with interconnected macroporous networks.66 By balancing salting-out (Cit<sup>3-</sup>) and salting-in (Ca<sup>2+</sup>) agents, PGCNG self-assembled into a uniformly porous structure without energy-intensive freezing steps (Fig. 5a).66 The synergistic interplay of Ca2+-induced polymer chain dispersion and Cit<sup>3</sup>-driven nanofiber aggregation created a porous framework (Fig. 5b), which showed weak interfacial hydration and instant droplet absorption (Fig. 5c and d).66 This architecture endowed PGCNG with rapid water transport (36 g min<sup>-1</sup>, four orders faster than PGNG; shorter half swelling time) and superior water storage capacity (saturated water content: 10.1 g g<sup>-1</sup>), attributed to free water dominance and interconnected micropores (Fig. 5e).66 The optimized pore hierarchy governed by salting-out effects further enabled the gel to achieve an outstanding evaporation rate of 3.5 kg m<sup>-2</sup> h<sup>-1</sup> and an evaporation efficiency of 93%.66

While Hofmeister effect-enhanced hydration significantly improves salt rejection and evaporation efficiency, the long-term durability of PVA-based hydrogels under cyclic mechanical stress remains a practical challenge. Hydrated polymer networks are usually fragile and prone to structural fatigue under repeated swelling/contraction or water flow impact.<sup>77-79</sup> To ensure the durability of these excellent hydration-related chemical properties, researchers have explored a variety of strengthening strategies: (1) constructing a dual network structure, such as a PVA/polyacrylamide (PAM) interpenetrating network, which uses a covalently crosslinked PAM network to provide a rigid backbone while preserving ion responsiveness;<sup>73,74,76</sup> (2) the introduction of nano-reinforced phases such as GO or cellulose nanofibers (CNFs) to improve the overall

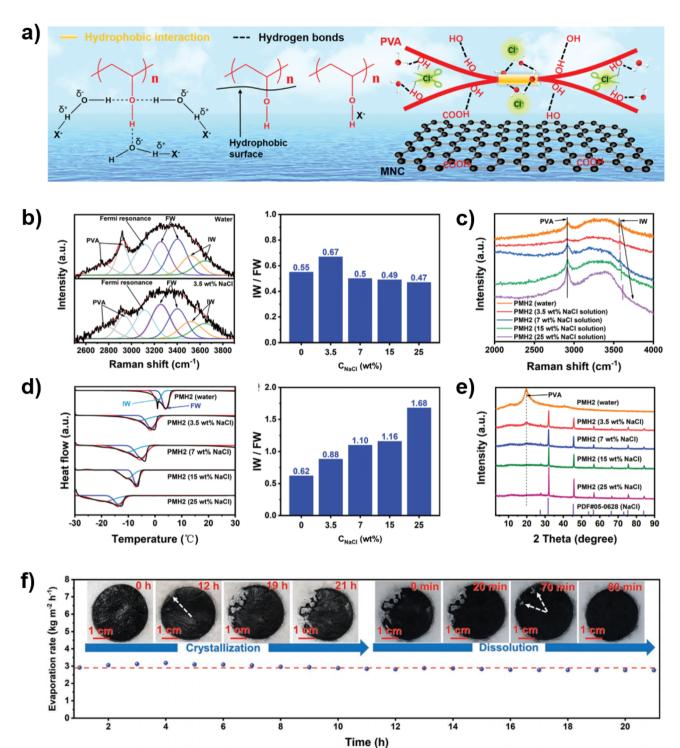


Fig. 3 Hofmeister effect driven hydration of PMH gels for solar-driven evaporation. (a) Schematic illustrations of interactions among X-, PVA, water, and salt ions within PMH. (b) Raman spectrum of the PMH swollen in water and 3.5 wt% NaCl solution; corresponding IW/FW ratios. (c) Comparative Raman spectra of PMH swollen in brines of varying NaCl concentrations. (d) Differential scanning calorimetry (DSC) curves of PMH swollen in brines of varying NaCl concentrations and the corresponding IW/FW ratios. (e) X-ray diffraction (XRD) patterns of PMH after swelling in brines of varying NaCl concentrations. (f) Evaporation rate of PMH in 3.5 wt% NaCl solution, with photographic insets demonstrating salt accumulation and re-dissolution on the evaporative surface. Reproduced from ref. 28 with permission from Wiley-VCH, copyright 2023.

mechanical properties;<sup>32,80</sup> (3) phase separation or cross-linking density regulation induced by salt ions (*e.g.*, Ca<sup>2+</sup>, Cit<sup>3-</sup>) is also used to optimize the network topology to take into account both

hydration capacity and toughness.<sup>66,70</sup> The long-term evaporation performance of these hydrogel evaporators is summarized in Table 1. For example, Yu *et al.* reported that PVA/PPy dual-

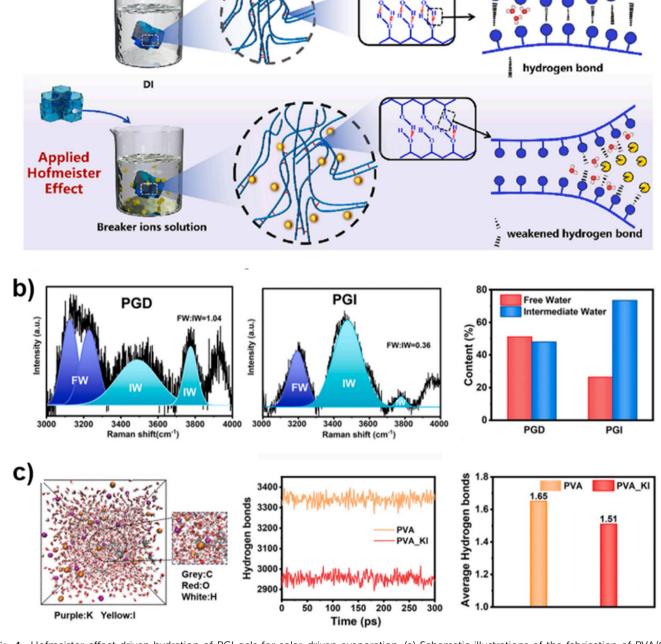


Fig. 4 Hofmeister effect driven hydration of PGI gels for solar-driven evaporation. (a) Schematic illustrations of the fabrication of PVA/GO aerogel, PGD hydrogel and PGI hydrogel. (b) Raman spectrum of the PGD and PGI hydrogels, with corresponding IW/FW ratios and the content of FW and IW in PGD and PGI. (c) Hydrogen bonding dynamics in PVA-KI systems: molecular simulation model with total and per-watermolecule hydrogen bond counts. Reproduced from ref. 32 with permission from Elsevier, copyright 2023.

network hydrogels could maintain evaporation rates of up to 3.2 kg m<sup>-2</sup> h<sup>-1</sup> after 672 hours of continuous working in seawater, demonstrating excellent long-term stability.8 These mechanical enhancement strategies are key to ensuring the long-term application of Hofmeister effect-driven hydration in actual desalination scenarios.

#### 3.2 Zwitterion polymers for solar-driven water evaporation

3.2.1 Characteristics and anti-polyelectrolyte effect of zwitterionic polymers. Zwitterionic polymers refer to polymers that contain equal amounts of anionic and cationic groups in repeating units; their positive and negative charges are uniformly distributed at the molecular scale.81 Fig. 6 presents the chemical structures of common zwitterionic polymers. The

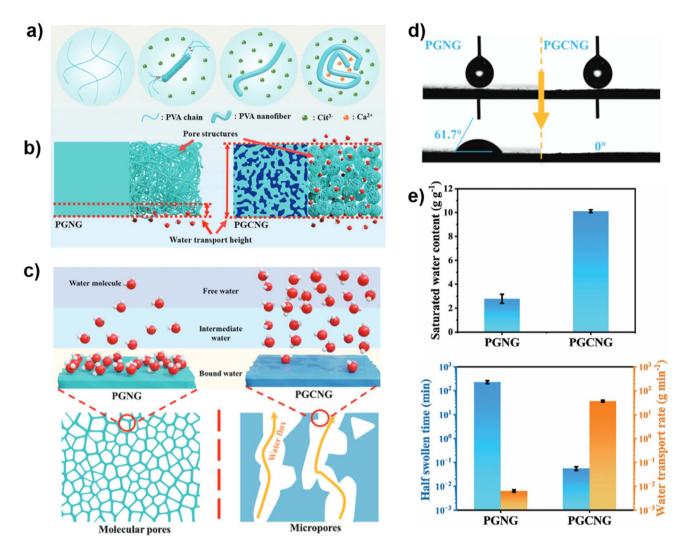


Fig. 5 Hofmeister effect regulated pore structure in PGCNG gels for solar-driven water evaporation. (a) Schematic illustrating the configuration of PVA chains and nanofibers. (b) Schematic illustrations of water uptake and transport in PGNG and PGCNG gels. (c) Schematic diagrams of the different water states (bound water/IW/FW) and comparative pore architecture (size/interface area) in PGNG vs. PGCNG gels. (d) Water contact angle photographs of PGNG and PGCNG gels. (e) Saturated water content, half swelling time and the calculated water transport rate of PGNG and PGCNG gels. Reproduced from ref. 66 with permission from Wiley-VCH, copyright 2024.

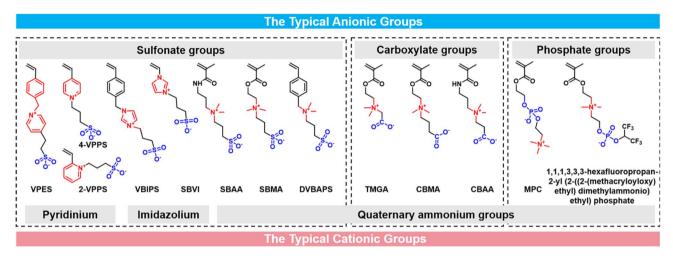
common cationic groups include quaternary ammonium groups (-N<sup>+</sup>R<sub>3</sub>), imidazolium groups and pyridinium groups, while the typical anionic groups are sulfonate groups (-SO<sub>3</sub><sup>-</sup>), carboxylate groups (-COO<sup>-</sup>), and phosphate groups (-OPO<sub>3</sub><sup>-</sup>).82 Currently, the most extensively studied zwitterionic polymers are polycarboxybetaine (PCB), polysulfobetaine (PSB), polysulfatobetaine, and polyphosphatidylcholine.82 Zwitterionic polymers possess an extremely high density of ionic groups, which enables strong interactions between the molecular chains and water molecules. This high-density ionic groups result in the formation of a dense hydrated layer on the surface of the material, thereby imparting high hydration properties.83-85 Besides, zwitterionic polymers exhibit an antipolyelectrolyte effect in brine: the salt ions in brine can shield the electrostatic associations between zwitterionic polymer chains, leading to the expansion of networks. Ultimately, this

further enhances the hydration capacity and the brine and solute transport ability of zwitterionic polymer hydrogels.<sup>86-91</sup>

It is worth noting that the degree of anti-polyelectrolyte effect is highly dependent on the molecular structures of zwitterionic moieties. Generally, a significant anti-polyelectrolyte effect lies in: (1) strong zwitterion–zwitterion binding energy to form a dense and compact cross-linked network, which provides sufficient space for subsequent salt-responsive swelling; (2) higher zwitterion-salt binding energy compared to zwitterion–zwitterion interactions, ensuring the screening of inter-chain interactions to release hydrated groups. Generally, the binding energy of sulfonate groups (-SO<sub>3</sub><sup>-</sup>) is stronger than that of carboxylate (-COO<sup>-</sup>) groups, and inclusion of aromatic structures (*e.g.*, benzene rings, imidazole and pyridinium) can further enhance the electrostatic interactions. As a result, the anti-polyelectrolyte effect of PSB is significantly stronger than

Table 1 A summary of the long-term evaporation performance of PVA-based hydrogel evaporators

Hydrogel name	Runtime	Evaporation performance	
PPy/PVA gel	672 h	Performance retention ratio: 99%, evaporation rate: 3.2 kg m <sup>-2</sup> h <sup>-1</sup> in seawater	8
PVA/PSS gel PVA/JGO hybrid gel	168 h 168 h	Performance retention ratio: 98%, evaporation rate: 3.6 kg m <sup><math>^{-2}</math></sup> h <sup><math>^{-1}</math></sup> in seawater Performance retention ratio: 98%, evaporation rate: 4.1 kg m <sup><math>^{-2}</math></sup> h <sup><math>^{-1}</math></sup> in 3.5 wt% saline water	73 80
PVA/PTA-PH1 gel	120 h	Performance retention ratio: 97%, evaporation rate: 3.9 kg m <sup>-2</sup> h <sup>-1</sup> in seawater	76
PVA/PAM gel	84 h	Performance retention ratio: 97%, evaporation rate: $4.0 \text{ kg m}^{-2} \text{ h}^{-1}$ in 10 wt% saline water	74



Chemical structures of common zwitterionic polymers.

that of PCB, and the introduction of aromatic structures will further amplify the anti-polyelectrolyte effects.92

In addition to the molecular structure of zwitterionic polymers, the type of salt ion also significantly influences its antipolyelectrolyte effect.<sup>93</sup> (1) For cations: divalent cations (e.g., Ca<sup>2+</sup>, Al<sup>3+</sup>) exhibit stronger binding affinity to anionic groups in zwitterionic polymers (e.g., sulfonate groups, -SO<sub>3</sub><sup>-</sup>) compared to monovalent cations. Increased interaction can disrupt interchain electrostatic interactions more effectively, thereby triggering a stronger anti-polyelectrolyte effect. 93 (2) For anions: the impact aligns with the Hofmeister series: the kosmotropic ions (e.g., SO<sub>4</sub><sup>2-</sup>) can induce a collapsed polymer chain state and reduced hydration capacity, while the chaotropic ions (e.g., Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, Br<sup>-</sup>) can promote a pronounced anti-polyelectrolyte effect (e.g., chain expansion) and enhanced hydration capacity by forming strong zwitterionic polymer-salt pairs with high binding energy, which provides enough driving force to dissociate more original inter-chain electrostatic associations.93

Such a salt-induced hydration phenomenon not only results in greater water uptake and faster water transport in highsalinity brines, but also constructs a dense hydration barrier against biofouling agents (e.g., proteins, bacteria). 92,94-96 By synergizing hydration enhancement with antifouling properties, polyzwitterionic hydrogels address both efficiency and durability challenges in solar-driven water evaporation.

3.2.2 Polyzwitterion-based materials for solar-driven water evaporation. Zwitterionic polymers not only achieve ultra-strong hydration through ionic hydration, but significantly enhance salt resistance through anti-polyelectrolyte effects, ultimately imparting strong intrinsic salt tolerance to evaporators.73,89-91 Yu et al. developed a polyzwitterionic hydrogel evaporator based on PSB, named as PZH.97 Raman spectra were used to evaluate the region of O-H stretching to analyze the hydrogen bonding distinction of water molecules within the PZH hydrogel.97 The peaks at 3233 cm<sup>-1</sup> and 3401 cm<sup>-1</sup> were referenced to FW, while the peaks at 3514 cm<sup>-1</sup> and 3630 cm<sup>-1</sup> corresponded to IW.<sup>97</sup> In virtue of the anti-polyelectrolyte effects of PSB, the polymer networks greatly activated the water molecules in concentrated brine, leading to an increased amount of IW and reduced evaporation enthalpy (817.4 J g<sup>-1</sup>) (Fig. 7a).97 Additionally, such saltinduced hydration phenomenon results in greater water uptake in high-salinity water, ultimately endowing the device with higher resistance to salt accumulation. Fig. 7b shows that the salt crystal (NaCl) could be eliminated within 30 minutes on the PZH hydrogel, highlighting its salt rejecting ability.97

Besides water management, the strong hydration capacity of zwitterionic polymers enable them to form a tightly bound hydration layer, thus effectively hindering the adsorption of contaminants like proteins, algae and bacteria, thereby endowing the device with excellent antifouling performance to ensure long-term operation. 30,31,98-102 The anti-biofouling efficacy of the composite polyzwitterionic hydrogel, driven by the PSB segments, was validated through bacterial adhesion and surface contamination experiments (Fig. 7c and d).30 As shown in Fig. 7c, the optical density (OD<sub>600 nm</sub>) values of Staphylococcus aureus (S. aureus) and Escherichia coli (E. coli) suspensions after cocultivation with hydrogels exhibited a significant inverse correlation with the PSB content.30 The results

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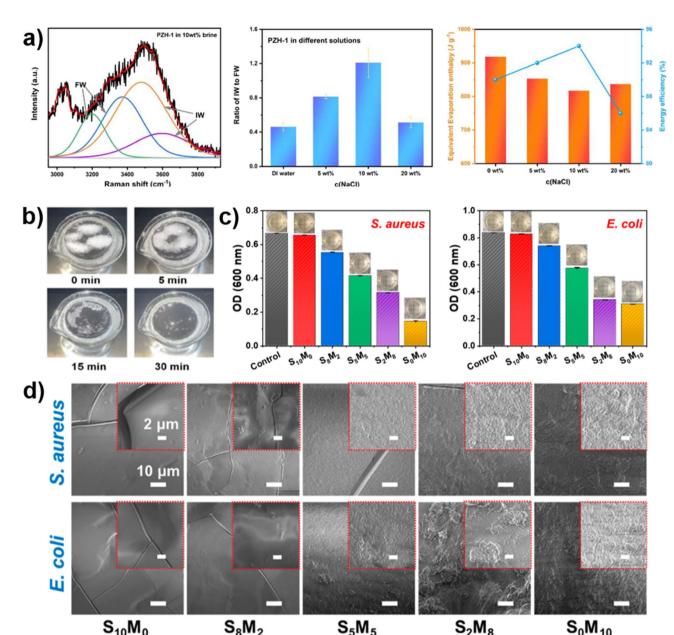


Fig. 7 Anti-polyelectrolyte effect driven hydration of PSB gels for solar-driven water evaporation. (a) Raman spectra, IW/FW ratios, the associated equivalent evaporation enthalpy and corresponding energy efficiency of PZH gels in brines with different concentrations.<sup>97</sup> (b) Saltresistant evaporation performance of PZH gels with pre-deposited salt crystals under 1 sun.<sup>97</sup> Reproduced from ref. 97 with permission from Wiley-VCH, copyright 2022. (c) Anti-fouling assessment: *S. aureus* and *E. coli* colony morphologies (photographs) and enumeration data.<sup>30</sup> (d) SEM images of the surfaces of  $S_{10}M_0$ ,  $S_8M_2$ ,  $S_5M_5$ ,  $S_2M_8$ , and  $S_0M_{10}$  hydrogels after cocultivation with insets highlighting bacterial distribution.<sup>30</sup> Reproduced from ref. 30 with permission from American Chemical Society, copyright 2022.

highlighted the PSB-dominated suppression of bacterial adhesion.<sup>30</sup> Similarly, SEM imaging (Fig. 7d) revealed a stark reduction in bacterial colonization on hydrogel surfaces with higher SBMA ratios.<sup>30</sup> This exceptional anti-biofouling performance is attributed to the anti-polyelectrolyte effect intrinsic to polyzwitterionic PSB.<sup>30</sup>

Furthermore, Zheng and Yang *et al.* synthesized two specifically designed sulfobetaine [dimethyl-(4-vinylphenyl) ammonium propane sulfonate (DVBAPS) and 3-(1-(4-vinylbenzyl)-1*H*-imidazol-3-ium-3-yl) propane-1-sulfonate (VBIPS)] zwitterionic gels; the relationship between the anti-polyelectrolyte effect and

molecular structure was studied (Fig. 8a).<sup>92</sup> The results showed the interaction of sulphonate groups is stronger than that of phosphates and carboxylates, and the introduction of benzene rings or imidazole will further enhance the anti-polyelectrolyte effect.<sup>92</sup> Fig. 8b and c collectively highlighted the water state regulation and water evaporation enthalpy of the Pvbips hydrogel. The Raman spectrum, the DSC profiles and the MD simulation results revealed a significantly higher IW/FW ratio in Pvbips (1.58 in 10 wt% brine) compared to Psbma (0.80), attributed to the strengthened water binding ability of the Pvbips network in brine.<sup>92</sup> Such activated water resulted in the

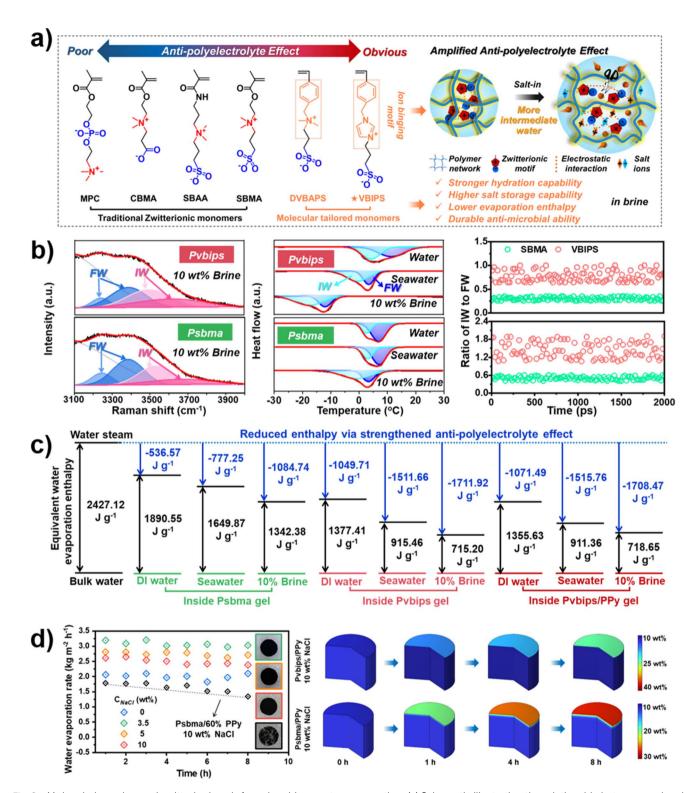


Fig. 8 Molecularly engineered zwitterionic gels for solar-driven water evaporation. (a) Schematic illustrating the relationship between molecular structure and anti-polyelectrolyte effect. (b) Evaluation of the IW/FW ratios of Psbma and Pvbips gels in different aqueous environments through Raman spectroscopy, DSC and MD simulations. (c) The calculated equivalent water evaporation enthalpy of different gels that were swollen in diverse aqueous environments. (d) Long-term evaporation performance of the Pvbips/PPy gel in saline water, with Psbma/PPy gel for comparison, with insets showing the salt precipitation condition of gels after 8 h of evaporation; salinity concentration simulation of Pvbips/PPy and Psbma/PPy gel after 8 h of evaporation in 10 wt% brine. Reproduced from ref. 92 with permission from Wiley-VCH, copyright 2023.

Table 2 A summary of the long-term evaporation performance of polyzwitterion-based hydrogel evaporators

Hydrogel name	Runtime	Evaporation performance	Ref.
P(SBMA-co-METAC) gel	1080 h	Performance retention ratio: 99%, evaporation rate: 1.6 kg m $^{-2}$ h $^{-1}$ in seawater Performance retention ratio: 98%, evaporation rate: 4.1 kg m $^{-2}$ h $^{-1}$ in 10 wt% saline water Performance retention ratio: 80%, evaporation rate: 4.5 kg m $^{-2}$ h $^{-1}$ in 10 wt% saline water Performance retention ratio: 97%, evaporation rate: 3.5 kg m $^{-2}$ h $^{-1}$ in pure water	30
DMAPS gel	504 h		97
PSBMA/ZIF-8/CNT gel	240 h		99
PDMAPS/PNIPAM gel	168 h		91

Table 3 A summary of hydrogel based evaporators with salt-triggered hydration character

Hydrogel name	Hydration mechanism	Evaporation performance	Ref.
PVA/modified needle coke gel	Hofmeister effect	Evaporation enthalpy: 1123 J g <sup>-1</sup> , evaporation rate: 3.18 kg m <sup>-2</sup> h <sup>-1</sup> in	28
PVA/graphene oxide/iodide gel	Hofmeister effect	3.5 wt% NaCl solution Evaporation enthalpy: 643.5 J g <sup>-1</sup> , evaporation rate: $\sim$ 2.1 kg m <sup>-2</sup> h <sup>-1</sup>	32
Carbonized chitosan gel	Hofmeister effect	in 3.5 wt% NaCl solution Evaporation enthalpy: 1397 J g <sup>-1</sup> , evaporation rate: 2.38 kg m <sup>-2</sup> h <sup>-1</sup> in	60
PVA/PAM gel	Hofmeister effect	seawater Evaporation enthalpy: 996 J g <sup>-1</sup> , evaporation rate of 4.0 kg m <sup>-2</sup> h <sup>-1</sup> during 16-h evaporation in 5 and	74
Pvbips/PPy gel	Anti-polyelectrolyte effect	10 wt% saline water Evaporation enthalpy: 911.4 J g $^{-1}$ , evaporation rate of 3.17 kg m $^{-2}$ h $^{-1}$ during 8-h evaporation in 3.5 wt%	92
DMAPS gel	Anti-polyelectrolyte effect	saline water Evaporation enthalpy: 817.4 J g $^{-1}$ , evaporation rate of 4.14 kg m $^{-2}$ h $^{-1}$ in 10 wt% saline water	97

ultra-low evaporation enthalpy of water in Pvbips gel (715.20 J g $^{-1}$  in 10 wt% brine), nearly half that of Psbma gel (1890.55 J g $^{-1}$ ). Perived from the strengthened anti-polyelectrolyte effect, the Pvbips/PPy gel maintained a stable evaporation rate of 3.17 kg m $^{-2}$  h $^{-1}$  in seawater after 8 h of desalination in seawater, outperforming the Psbma/PPy gel (declining from 1.78 to 1.34 kg m $^{-2}$  h $^{-1}$ ). Moreover, simulations revealed that Pvbips/PPy suppressed surface salinity accumulation below saturation levels, whereas Psbma/PPy failed, owing to the strengthened anti-polyelectrolyte effect (Fig. 8d). Period of the strengthened anti-polyelectrolyte effect (Fig. 8d).

Besides zwitterionic polymers, hydrogels based on oppositely charged polyelectrolytes also exhibit similar salt responsiveness. 103 Specifically, salt ions bind with the polyelectrolytes and thus screen the electrostatic interactions between polyanions and polycations, enabling network expansion and enhanced water uptake.103 For example, Zhu and Wu et al. developed a polyion complex hydrogel with hierarchical porosity, achieving an evaporation rate of 2.79 kg m<sup>-2</sup> h<sup>-1</sup> in 3.5 wt% NaCl solution under 1 sun irradiation, surpassing its performance in pure water. 90 In this case, the electrostatic pairs poly(sodium styrenesulfonate) polv(3methacryloylamino)propyl-trimethylammonium chloride dissociate in brine, leading to the increase of water adsorption and decrease of evaporation enthalpy.90 Sharing a similar swelling behavior with zwitterionic hydrogels, these systems

differ in requiring precise charge balance and exhibiting more sensitive pH-dependent stability.<sup>81,96,104</sup>

Despite the exceptional anti-fouling and salt-resistant ability of polyzwitterionic hydrogels, their low mechanical strength limits service life in practical applications.<sup>81</sup> Hybrid designs combining zwitterionic polymers with double-network structures (*e.g.*, alginate-polyzwitterion) can achieve high fracture energy, enabling resilience against wave erosion in marine environments.<sup>30,91,94,102</sup> Therefore, combining the advantages of salt-triggered hydration with the intrinsic nature of the material and the targeted mechanical strengthening design is an effective way to develop a practical and durable zwitterionic evaporator. The overall long-term evaporation performance of these polyzwitterion-based evaporators is summarized in Table 2.

In summary, salt ion triggered hydration can improve the brine pumping ability of evaporators and reduce the evaporation enthalpy of water even in highly concentrated brine environments. These points are the key working mechanisms of these salt rejecting hydrogels. The overall evaporation performance of these hydrogel evaporators is summarized in Table 3.

### Conclusion and outlook

The rapid development of hydrogel-based solar-driven desalination technology has opened up new avenues for addressing global freshwater scarcity. This review highlights the critical role of hydration chemistry in optimizing hydrogel materials for high-efficiency solar-driven water evaporation, with a focus on two pivotal strategies: the Hofmeister effect in PVA-based materials and the anti-polyelectrolyte effects of zwitterionic materials. These approaches have demonstrated remarkable potential in overcoming the limitations of conventional desalination methods, such as high energy consumption and salt fouling through tailoring water transport pathways, reducing evaporation enthalpy, and enhancing salt tolerance. Hydration chemistry has emerged as a cornerstone for advancing solar-driven desalination. By bridging hydration chemistry with desalination technology, hydrogel-based solar-driven water evaporation systems offer a sustainable pathway to decentralized water purification, aligning with global decarbonization and water security goals.

However, the transition from laboratory breakthroughs to real applications faces many challenges<sup>105–108</sup> and needs further investigation:

- (1) The hydrated polymer network is generally fragile and weak, restricting the service life of these hydrogel based evaporators. Developing hydrogels with a combination of high hydration and mechanical strength needs more complicated design of network topology.
- (2) Beyond desalination, salt-triggered hydration hydrogels hold potential in ion selective desalination and mineral resource recovery. These hydrogels might preferentially bind to certain ions, which facilitates concurrent salt rejection and mineral concentration. The ion-specific hydration mechanisms might provide new pathways for the simultaneous collection of high-value minerals (*e.g.*, Li<sup>+</sup>, Mg<sup>2+</sup> salts) from high-salinity brines or wastewater, significantly enhancing techno-economic viability. However, more complicated molecular design of the hydrogel is needed to strengthen its ion selectivity. Besides, further synergistic coupling with the design of evaporation interfaces (*e.g.*, Janus evaporator-crystallizer designs) is also required.
- (3) Water-electricity co-generation represents a transformative opportunity for maximizing resource utilization: evaporation-induced thermal gradients or salinity differences might drive simultaneous electricity generation. However, the effect of salt-triggered hydration on electricity generation is still unclear.
- (4) Salt induced hydration chemistry provides a new avenue to promote evaporation speed in highly concentrated saline water. Therefore, the water collection systems should also be effective enough to match the speed of steam generation.

### Conflicts of interest

The authors declare no conflict of interest.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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