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Abstract: The safe and economical management of shale gas flowback and produced water (FPW) with the goal of zero-liquid discharge (ZLD) is of paramount significance to the sustainable development of the energy industry. This challenge is still widely impending, due to severe limitations related to the complexity of FPW streams and high costs associated with their treatment. A long-term feasible solution is represented by solar-driven interfacial evaporation (SIE), a low-cost and environmentally friendly desalination technology. Technical and economic analyses show that even in the Sichuan Basin, where solar intensity is low and the volume of FPW is large, 4000 m^2 of solar still would be sufficient to accomplish ZLD with a capital cost lower than \$ 1 m^{-3}, significantly cheaper than traditional membrane-based and thermal-based technologies (with costs above \$ 15 m^{-3}). Beneficial products, including condensate water and crystalline salts, may also be effectively recovered in this process, although design improvements are needed in this area. This study also discusses possible solutions to address the passage of volatile organic compounds into the effluent and to achieve the smart recovery of strategic resources, such as lithium and rare earth elements. The major current challenges of solar-driven evaporation for the beneficial management of FPW are scaling and fouling, cost-effective latent heat recovery, the scalability of the systems, and efficient water production and salts harvesting. These issues require ad hoc research efforts, analyses, and pilot testing.

Keywords: Solar-driven interfacial evaporation; flowback and produced water; desalination; zero-liquid discharge; resources recovery

1. Introduction

Extraction of unconventional shale oil and gas has profoundly affected the energy landscape worldwide, particularly in the United States and China [1]. However, horizontal drilling and hydraulic fracturing (HF) techniques rely heavily on water consumption, as between 20,000 and 60,000 m^3 water is needed for the exploitation of each well, according to a recent research report [2]. The total water consumption has been steadily growing in most shale gas regions, partly because an increasing number of wells are drilled and exploited, but also due to the increasing flow of water required for each well [3]. During the extraction activities, a percentage of HF water in the range of 5-85% re-surfaces and generates large volumes of shale gas flowback and produced water (FPW) [4, 5]. According to Rystad Energy (2019), over 1.2 billion m^3 of water will be demanded for HF operations in the U. S. in 2021, thus producing a staggering amount of shale gas FPW. An additional challenge is that the typical physicochemical composition of shale gas FPW is greatly complex, mainly including high levels of total dissolved solids (TDS), a variety of hazardous organic compounds, and naturally occurring radioactive materials (NORM) [6]. Therefore, the effective management of shale gas wastewater is not only related to the sustainable development of the shale gas industry, but also of great significance to the health of residents and water environments in shale regions [7, 8].

The reuse of shale gas FPW has attracted increasing attentions in recent years to help improving the sustainable operation of the energy industry [9-11]. Recycling FPW for the fracking of subsequent wells is defined as internal (or on-site) reuse, a technique that reduces the demand of freshwater withdrawal and that is cost-effective because the aqueous stream does not require desalination. However, internal reuse is a temporary solution, because the FPW quality would gradually deteriorate with increased reuse cycles, in turn reducing natural gas production

[12]. Additionally, the volume of FPW may exceed the internal reuse capacity, and opportunity for internal reuse would eventually vanish. Therefore, FPW would eventually require advanced and complex treatment to remove organic components and salinity and to reach quality levels suitable for other types of beneficial external reuse (*e.g.*, irrigation, livestock watering, aquifer recharge) or for safe discharge in surface water bodies [6]. Mechanical vapor compression (MVC) and membrane-based technologies have been proposed and studied for the desalination of FPW, but these methods have not been used at large scale and are restricted by economic and/or technological limitations [13].

Solar-driven interfacial evaporation (SIE) technology realizes desalination by exploiting renewable energy [14, 15]. It is regarded among the most practical and economical solar-thermal desalination technologies [16]. SIE has been already applied to obtain potable water from seawater or surface waters by portable solar stills [17, 18]. Novel applications of this method also include salt extraction, wastewater zero-liquid discharge (ZLD), and electricity generation [19]. In recent years, advancements in solar absorber materials and system designs for SIE have greatly improved the solar-to-steam conversion efficiency [19-22], and various relatively large outdoor solar stills have been developed and employed for saline water desalination [23], paving the way for its large-scale industrial production. SIE for desalination is not restricted by water salinity levels and can extract high-quality water all the way to crystallization [21]. Therefore, SIE may be a feasible technology for the desalination of shale gas FPW with the goal of achieving minimal and zero-liquid discharge. The condensate water and the precipitated salts from this process may also be recovered. The most attractive means to accomplish this goal is arguably the recycling of strategic resources, namely, lithium (Li) and rare earth elements (REEs), which exist at varying concentrations in shale gas FPW [24-26]. Until now, no research has addressed the implementation of SIE for shale oil and gas wastewater treatment, reuse, and valorization, which however presents remarkable opportunities for promoting the sustainability of shale oil and gas production. Pursuing such opportunities requires in-depth and systematic understanding of the potential and challenges to guide the research, development, and implementation of SIE for the sustainable management of shale oil and gas wastewater.

2. Challenges in treatment and reuse of global shale gas FPW

Salinity, typically quantified by total dissolved solids (TDS), is an important index for the design of a treatment sequence and for the identification of the most appropriate desalination method. The salinity varies greatly for different shale gas plays. In the U.S., the reported TDS values for FPW range from 12,000 to 300,000 mg/L [4, 9]. In the Sichuan Basin in China, the TDS value is commonly lower than that reported in the US and below 40,000 mg/L [6, 27]. For a specific shale gas well, the wastewater volume and quality also vary temporally in different stages of utilization, namely, the flowback period, the transition period, and the produced water period [28], with the salinity continuously increasing along the well lifetime [29]. Large regional differences in salinity and continuous changes over time complicate the selection and design of treatment methods, because each technological configuration functions efficiently within a certain salinity range and for a relatively constant feed water composition. For example, reverse osmosis (RO) has a salinity limit of roughly 70,000 mg/L for the feed water [13, 30]. Other thermal desalination technologies, such as mechanical vapor compression (MVC), are more suitable for hypersaline wastewaters but are generally more costly [13].

In the U.S., three strategies are employed to manage shale gas FPW: i) deep well injection; ii) internal (on-site) reuse; iii) external reuse or disposal after treatment. Internal reuse is only a temporary solution because recycled wastewater keeps flowing back to the surface and eventually this stream will require a desalination treatment or ultimate disposal through deep well injection. Deep well injection is relatively inexpensive, which is the main reason why several shale plays in Texas, including Barnett Shale, Eagle Ford Shale, and Haynesville Shale apply this technique, especially considering the availability of deep wells in those locations [31]. However, deep well injection raises growing public concerns due to its risks in contaminating groundwater aquifer [4] and inducing earthquakes [32]. In the Marcellus Shale in Pennsylvania, 74% of the FPW is reused for subsequent HF activities after partial treatment, while 16.2% of the flow is externally reused after desalination. The fraction of wastewater injected into disposal wells has recently fallen under 10% [33, 34].

In China, the Sichuan Basin is currently the most productive basin for shale gas exploitation, and has entered a commercial development stage. Fuling, Weiyuan, Changning, and Zhaotong are the four major shale gas production regions [35]. In these regions, most of the shale gas FPW (> 85%) is reused on site for further HF with simple treatment or without any treatment, while the remaining fraction is stored in ponds or injected into deep wells. However, some desalination treatment technologies are under development in recent years. The Fuling shale gas field is at the forefront of this development. The FPW in Fuling cannot be injected into deep wells due to restrictions related to adverse geological conditions, therefore, desalination treatment is urgently needed. In 2020, an FPW treatment plant with a capacity of 1600 m^3/d was inaugurated in Fuling. A hybrid membrane-based ZLD treatment train is in place, including ultrafiltration (UF), RO, and MVC [36]. However, the treatment cost is over \$ 15 m^{-3} of wastewater. In addition to the high treatment cost, transporting the FPW from nearby wells to the treatment plant costs up to \$ 0.3 m^{-3} for each km. Treatment plants are also being built for the Changning field. However, restricted by unique topography, wells and platforms in Changning are usually far apart, rendering centralized treatment more expensive. Such high expenditures may be cost prohibitive for the shale gas industry to practice sustainable wastewater management.

As for Canada, very limited reports have discussed the water footprint of hydraulic fracturing in British Columbia (BC) [37-39], which hosts one of the world's largest reserves of recoverable shale gas, including four shale gas basins in the northeastern British Columbia (NEBC). These are the Montney, Horn River, Liard Basins, and the Cordova Embayment [37]. The work of Wisen et al. indicated that the TDS concentration in the shale gas FPW from the Horn and Montney basins is between 100 and 160 g/L. In NEBC, deep well injection is the only permitted method for the disposal of the shale gas FPW that is not re-used for subsequent reservoir stimulation operations, according to the British Columbia Oil and Gas Commission [38]. Wastewater cannot be released to surface water bodies. However, the increase in reservoir pressure generated by wastewater injected into deep wells has been associated with seismicity throughout Canada and the United States [40]. Analysis of the FracFocus database and freshwater withdrawal data from the Integrated Resource Information System (IRIS) database suggests that only approximately 40% of hydraulic fracturing fluids are exploited for internal reuse [38].

For the countries mentioned above, the fraction of shale gas FPW that is presently desalinated is very small, and the current management options are distant from being sustainable. Although scientists are developing several efficacious processes to treat shale gas FPW, most of these processes are characterized by low technology readiness level, i.e., these processes have been validated only at laboratory scale and are far from being sufficiently mature for field implementation. Despite the increased attention around minimal or zero liquid discharge, existing technologies capable of achieving this treatment goal are costly and energy-intensive

[41]. Can we achieve ZLD of shale gas FPW with low environmental impacts? Addressing this question is important for the environmental compatibility and the sustainable development of the shale oil and gas industry. In this paper, we propose SIE technology as a feasible option to treat FPW. In the following discussion, we analyze potential, feasibility, and challenges of SIE and we benchmark SIE against conventional desalination technologies.

3. Feasibility of SIE technology

3.1. Principles, structure, and recent developments of SIE technology

The basic principle of SIE technology for ZLD of FPW is that SIE exploits solar energy via photothermal conversion to heat water at the air-liquid interface to drive the evaporation of water and generate water vapor—a mechanism that improves the solar-to-vapor efficiency beyond 90% [16]. The vapor is then collected and condensed to obtain freshwater. This process can push the desalination to the limit of zero liquid discharge and obtain salt crystals as a by-product [42]. The SIE device consists of the following key components: a solar absorber that can efficiently absorb and convert the solar radiation into heat while allowing the vapor to permeate through the front face; a floating evaporation structure that can simultaneously maximize the evaporation rate and supply liquid to the heated region; a thermal insulator that can effectively reduce the loss of the converted solar-thermal energy to the bulk liquid; and a condensation cover to condense and collect freshwater, if desired [16, 21]. The schematic is depicted in **Fig. 2a**.

Significant work has been carried out with the goal to enhance the efficiency of vapor generation by improving the solar absorption and the heat conversion efficiency of solar absorber materials, as well as optimizing the system design to minimize parasitic heat losses [42, 43]. More details about this process and its improvements can be found in a number of comprehensive reviews [19-22]. Low-cost and simple systems have been designed and achieved satisfactory efficiencies [17, 44], providing opportunities for practical applications in potable water production and to achieve ZLD of wastewater.

Some references [9, 36, 37] reported that careful structural designs can exploit environmental energy (low air humidity, wind, or temperature) to enhance the performance of an interfacial solar vapor generation device to well above the theoretical limit of vapor output, usually assumed to be 100% in terms of solar-to-vapor energy transfer efficiency, under various light intensities. However, maximizing water collection from vapor is still an enormous challenge because if the condensate water is collected, the solar-to-vapor efficiency of such closed system cannot exceed the theoretical limit when no latent heat is recovered. Note that latent heat recovery is difficult to be practically implemented in small-scale SIE systems in consideration of high capital costs and energy inefficiencies [21, 45].

3.2. Economic and technical feasibility

We provide a global map of basins with shale reserves and of the solar horizontal irradiation in Fig. 1. There are many regions with abundant shale reserves that are also rich in solar resource, such as the Tarim Basin and the Zhunger Basin of China, as well as areas in the west and southwest of the United States, in Argentina, Algeria, Mexico, Australia. Taking Texas as an example, being this State active in shale gas exploration [31, 46] while simultaneously rich in solar energy resources, SIE has great application potential for shale gas FPW treatment. A number of solar-thermal seawater desalination demonstration plants have been implemented over the last decade in Texas [47], providing suitable reference for further development.

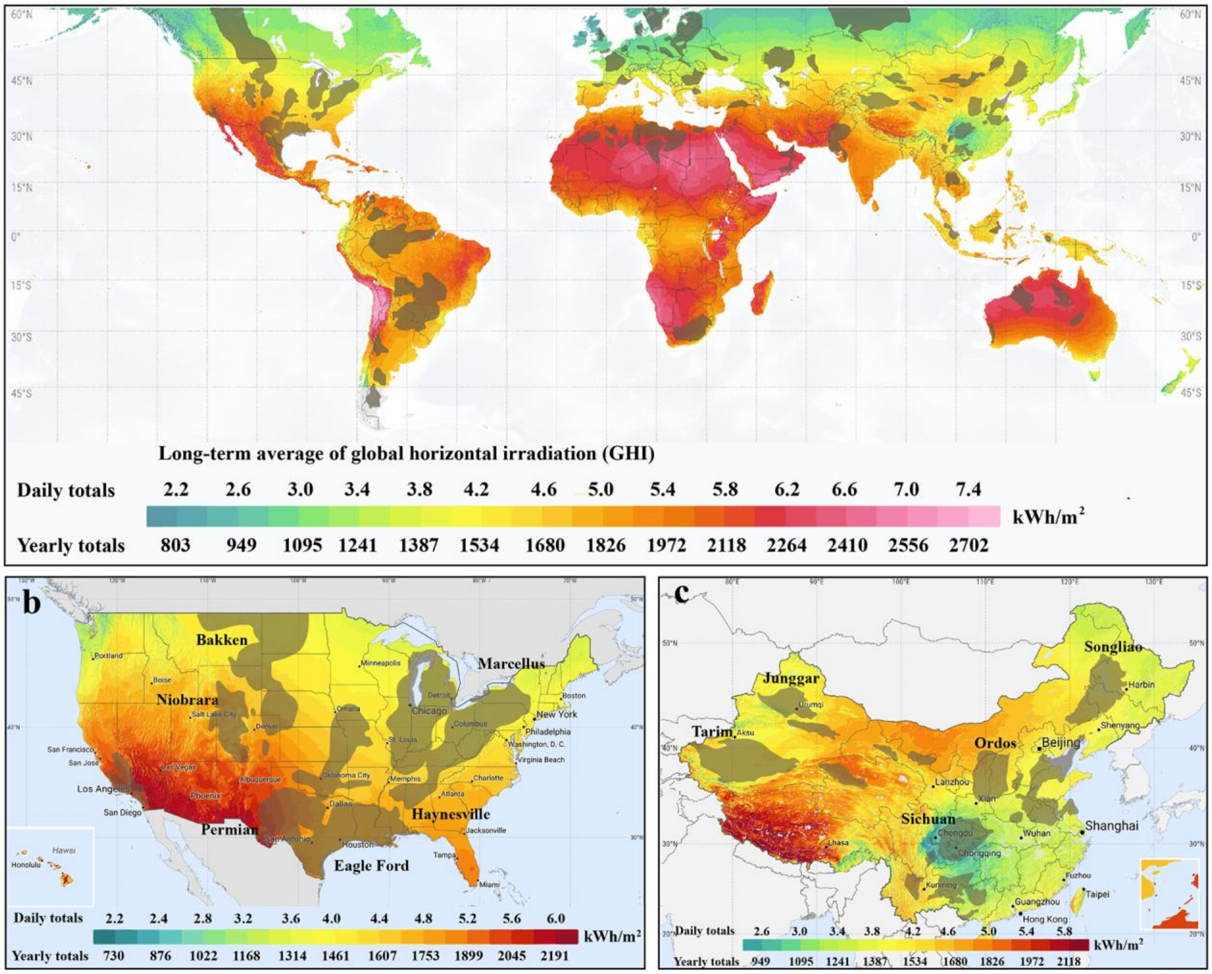

Fig. 1. Global map of solar resources and shale resources. (a) Map of global horizontal irradiation overlaid with the basins of shale reserves; data collected from Solargis (http://globalsolaratlas.info), the U.S. Energy Information Administration [48], and Geocloud [49] in China. Data on surface solar radiation are not shown in high-latitude regions, because few studies have evaluated their solar irradiation. (b) Solar horizontal irradiation and basins with shale resources in the U.S.[48] (c) Solar horizontal irradiation and basins with shale resources in China [49].

SIE technology has been deployed to collect clean water since ancient times [47], and now has mature applications in off-grid devices for water supply. Currently, countries like Australia, the

U.S., China, Chile, and Algeria have built solar stills with areas up to ~8000 m^2. Therefore, large-scale harvesting of solar energy is not technically restricted. However, this technology has rarely been used for large-scale desalination. This circumstance may be attributed to its inherent low specific water production (SWP) in closed system. When no latent heat is recovered, the maximum theoretical SWP value is 1.6 L m^{-2} h^{-1} under 1 sun (1 kW/m^2). And latent heat recovery is difficult to be implemented in SIE system, as mentioned above [21]. We assume that shale gas FPW is treated in such closed system without latent heat recovery, which is the most adverse case, to analyze the technical feasibility of SIE. Under this situation, a much larger area and much longer times are needed to treat the same amount of FWP compared with SIE configured as an open system or with latent heat recovery. As for wastewater volume, the largest fraction of shale gas FPW (80-90%) is typically reused on-site without desalination and would not require desalination, thus the typical flow of wastewater that needs treatment is not large. Although SIE will consume more time and larger land area than other desalination technologies, most shale plays are located in remote areas: time and land use are not critical constraints compared to other economic factors. This latter consideration is key to evaluating the feasibility of SIE for FPW treatment operations. We also assume that: i) one year is required for the treatment of FPW by SIE; ii) twenty percent of predicted FPW in 2030 will need to undergo desalination; iii) the SWP under one sun is 1 L m^{-2} h^{-1} (typically for solar stills), ignoring the losses and degradations of the evaporation process; iv) the SWP is equal to the rate of wastewater treatment. Based on these relatively conservative figures, the areas needed for solar stills in typical shale regions both in the U.S. and in China in 2030 are presented in Table 1. Taking Eagle Ford as an example, its theoretically predicted FPW volume in 2030 is 12000 m^3/well, according to Kondash et al. [3]. Twenty percent of predicted FPW will need to undergo desalination, which corresponds to 2400 m^3/well. The yearly total average horizontal irradiation of Eagle Ford is 2000 kWh/m^2, based on the data summarized in Fig. 1b. One sun is 1 kW/m^2, which means 2000 h irradiation time of one sun, yearly. The specific water productivity is roughly 1 L $m^{-2}h^{-1}$ under one sun, which means that 2000 L/m^2 FPW can be treated per square meter of solar still. Therefore, the solar still area needed for desalination in this location would be 1200 m^2.

Table 1. Calculation of the areas of solar stills needed to desalinate FPW in typical shale regions, in the U.S. (Eagle Ford, Haynesville, Marcellus and Permian) and in China (Weiyuan). The actual (2016) and theoretically predicted (2030) volumes of shale gas FPW in the U.S. refer to the work by Kondash et al.[3], the volumes related to China refer to the work by Zou et al.[27]

Shale gas region	FPW in 2016 (m^3/well)	Predicted FPW in 2030 (m^3/well)	Predicted FPW for desalination (m^3/well)	Yearly total average horizontal irradiation (kWh/m^2)	Specific water productivity (L $m^{-2}h^{-1}$)	Area of solar still (m^2)
Eagle Ford	8082	12000	2400	2000	1	1200
Haynesville	11509	17000	3400	1800	1	1900
Marcellus	2204	12000	2400	1400	1	1700
Permian	74471	300000	60000	2100	1	29000
Weiyuan	19800	NA[a]	4000[b]	1000	1	4000

Notes: [a] Not available; [b] This value is calculated using the volume of FPW in 2016 (19800 m^3) because that in 2030 is not available.

The results suggest that SIE is feasible in most regions when the goal is treating shale gas FPW, because the estimated solar still areas are within technical and economical reach. In the Sichuan Basin, each drilling platform has a pond covering an area of approximately 6000 m^2 to store FPW (Fig. 2b) [27], amply sufficient for solar still installation. The situation is similar in the U.S. Only for the Permian basin, the large volumes of FPW would require significantly longer treatment time and larger areas. In other cases, if the open system is adopted (not collecting condensate water), the solar-to-vapor rate can be significantly improved to over 10 kg $m^{-2}h^{-1}$, beyond the thermodynamic limit [50]. Alternatively, the latent heat recovery is realized in a vapor-to-water process, significantly increasing the SWP. Both methods can greatly reduce the area of the solar still and the treatment time, at the expense of increasing installation and possibly maintenance costs. However, these techniques are not yet sufficiently for practical application. In our economic analysis, neither case is considered, and we just disscuss the closed system with no latent heat recovery.

From the point of view of the process economy, SIE is arguably the most economical of all current desalination technologies, because it does not require any mechanical moving parts, does not require high pressure or vacuum operation, and it exploits sunlight as its sole energy source [50]. Moreover, the device requires simpler maintenance compared with membrane-based technologies and other thermal desalination technologies [51, 52]. The materials to produce solar stills are generally widely available and low-cost, *e. g.*, cellulose-based fabric, expanded polystyrene, polymer films [17]. Although studies [53, 54] have highlighted the importance of installing optical concentration for seawater desalination plants, this additional feature is unnecessary for shale gas FPW treatment, given that time and land are not a limitation, as highlighted above. We estimate that the cost of SIE devices is about \$ 3 m^{-2} with a projected life-cycle of 2 years, according to the work of Ni et al. [17]. Thus, the capital cost for shale gas FPW desalination would be lower than \$ 1 m^{-3}. Although labor and maintenance costs are not assessed in this work, a much lower total cost can be expected than extracting freshwater using current desalination technologies (over \$15 m^{-3}, as discussed above).

3.3. Comparison with alternative desalination technologies

Membrane-based desalination technologies, including nanofiltration (NF), RO, forward osmosis (FO), and thermal-based membrane distillation (MD), as well as MVC, have been extensively investigated for shale gas FPW treatment in recent years and each presents its own advantages and disadvantages [6, 9, 13]. NF and RO are pressure-driven processes with high energy efficiency [55]. Their technical difficulty is low, and the commercial application is very mature. However, the maximum salinity tolerance of these processes is typically 70,000 mg/L, which implies that only FPW below this salinity value can be treated. Also, these processes are limited in terms of recovery rate, *i.e.*, the fraction of freshwater that can be extracted from the high salinity feed stream, with RO usually operating at or below 60% recovery rate. Note that the recovery rate of seawater (~35,000 mg/L) desalination is usually around 50%. Therefore, a concentrated solution is always produced, together with a freshwater stream, the former needing additional extensive desalination to reach ZLD or complex management for its safe disposal.

The FO process consists of a first separation step in which the driving force is represented by an osmotic pressure difference across the semipermeable membrane, a process that can virtually manage a salinity as high as 200,000 mg/L. The energy consumption is small in this first separation step, with possibly lower fouling tendency than RO [56]. However, the major obstacle of FO-based desalination is the downstream recovery of the draw solution (separation of the freshwater from the draw solute), an operation with high costs and often associated with operational complexity depending on the nature of the draw solute [57]. Even when freshwater extraction is not the goal and draw solution does not require regeneration, its management is not straightforward and this process would require the continuous supply of new and often costly draw solute. This technology has not been yet applied for desalination at large scale.

MD relies on a process that combines membrane technology with thermal technology, and this technique is attracting enormous interest for the desalination of hypersaline shale gas FPW with TDS even higher than 200,000 mg/L. The cost of MD is potentially much lower than that of MVC, because low-grade thermal energy in shale gas fields can be utilized. Research progresses have been made in MD for shale gas FPW treatment [58-60], however, pilot tests are still at initial stages. The membranes deployed in MD are easily wetted by surfactants present in shale gas FPW, a phenomenon that severely deteriorates the quality of the effluent water. Membrane scaling and fouling represent additional challenges that should be urgently solved for the large-scale implementation of MD. Finally, MVC is a widely adopted and commercially mature thermal desalination technology, already demonstrated in seawater desalination and high-salinity wastewater treatment. MVC equipments for shale gas FPW treatment has also been put into use in Texas and in Sichuan Basin [46]. However, the Achilles' heel of this technology is the high cost of installation, operation, and maintenance, as well as high energy consumption, all translating into significantly higher costs with respect to membrane-based technologies.

A qualitative comparison between SIE and the technologies mentioned above is summarized in Fig. 2c [13, 45]. SIE has great potential advantages in terms of overall cost, and may be widely applicable in the treatment of shale gas FPW with different salinity levels and volumes. Importantly, it can push desalination all the way to the crystallization limit and help achieving ZLD. Although the energy efficiency and SWP of this technique are low, they are sufficient for the treatment of shale gas FPW and these problems are offset by the efficient exploitation of solar irradiation. Considering environmental impacts, the SIE process utilizes solar energy as the only energy input, and the raw materials and device systems are much simpler than those of membrane-based and thermal-based technologies [15, 17]. Therefore, greenhouse gases emissions from SIE are lower than those of high-temperature thermal-based desalination technologies, which demand electricity generation [56], and those of membrane-based technologies, which depend on polymer membrane fabrication and also require the management of brines [61]. The by-products of the SIE process include condensate water and salt crystals, which may be reused, thus improving the sustainability of the process life cycle. Therefore, SIE has arguably the lowest carbon footprint and environmental impacts among the current desalination technologies, and it helps reducing the dependence on fossil fuels.

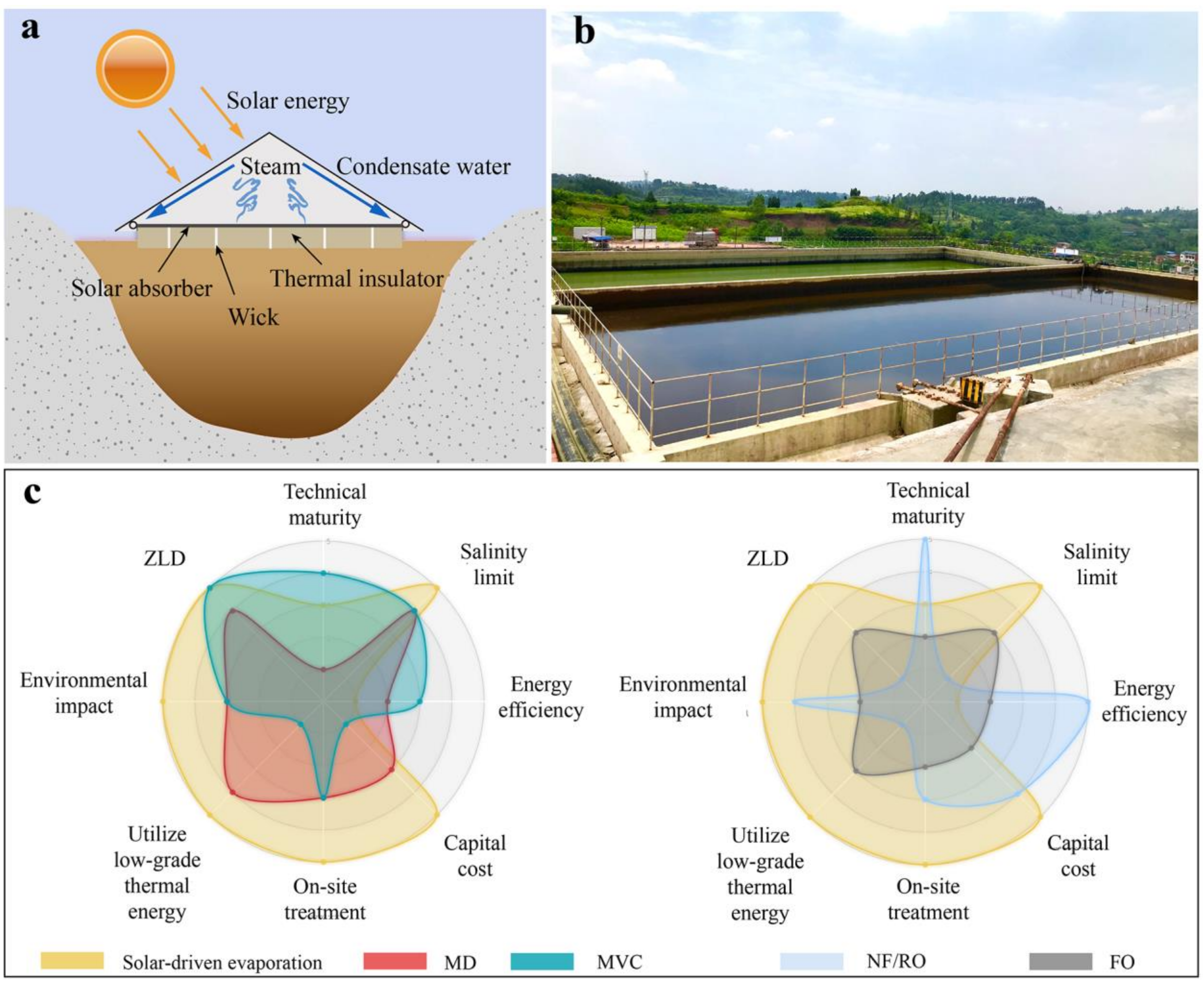

Fig. 2. Feasibility of SIE to treat shale gas FPW. (a) Schematic of the SIE system with a polymer-film based condensation cover floating on the FPW pond. (b) Photograph of a FPW pond in the Weiyuan shale play of the Sichuan Basin. (c) Qualitative comparison with other thermal-based and membrane-based desalination technologies for shale gas FPW treatment [13, 45].

4. Condensate water reuse

One simple configuration of the SIE device does not involve the collection of the condensate water, i.e., open system, letting instead the steam disperses into the surrounding air, which will further reduce the installation and management costs, and has the potential to reach much higher solar-to-vapor evaporation rate. However, taking into consideration water footprint and sustainability issues, collecting the condensate water for external reuse, *e.g.*, irrigation, may be the most appropriate choice in some cases. (Fig. 3e). It is reported that 38% of shale resources are in areas that are either arid or under high to very high levels of water stress. In China, this percentage is as high as 61%. Furthermore, 40% of shale plays are in areas of active agricultural activities, by far the largest water users [5]. The water quality standards for irrigation, livestock watering, and surface water discharge can be found in a previous review work [6]. Theoretically speaking, the quality of condensate water would be even adequate for drinking purposes; however, given the complexity and heterogeneity of FPW composition, analysis of the specific condensate water would be necessary to evaluate its quality, especially with respect to the presence of volatile organic compounds (VOCs) and semi-volatile organic compounds (sVOCs). VOCs and sVOCs have been observed in shale gas FPW in both the U.S. and China; they include benzene, toluene, ethylbenzene, and xylenes (BTEX), and other low molecular weight compounds [62, 63]. Previous studies have suggested that traditional thermal distillation technologies, *i.e.*, MD and MVC, will promote the passage of these contaminants into the gas phase, and then into the liquid water following condensation [46, 58]. Their concentration in the condensate water may even be enriched compared to the initial concentration in FPW by the distillation processes, resulting in levels beyond the safety limits [19, 60]. Research has been conducted also on SIE systems, reporting similar phenomena [64-67]. Shi et al. first found that solar-driven water evaporation will accelerate VOCs volatilization and enrich these compounds in the condensate [64]. Although no study has been carried out with shale gas FPW by SIE, we can expect analogous results. This process would greatly affect the management of the condensate water. Even if the condensate water is not collected for reuse, the steam being released to the atmosphere may pose environmental and human health risks related to VOCs and sVOCs contamination.

Several strategies are proposed to tackle this challenge:

i) Designing bifunctional solar evaporation systems. The inclusion of solar absorber materials possibly capable of catalytically degrade VOCs would efficiently remove these contaminants in-situ to avoid their accumulation in condensate water and their release into the environment [64, 65, 68]. For example, Ma et al.[65] have designed an MOF-based membrane with excellent photothermal properties and high Fenton catalytic activity to produce safe water from a stream contaminated with VOCs. Selective permeable membrane were also developed to enable the separation of safe water from VOC pollutants by solar evaporation, with a VOC removal rate of 90% [66]. Another idea is the deployment of tailored activated carbon to act both as solar absorber and as VOCs adsorber [67].

ii) Post-treatment of condensate water. Condensate water containing VOCs may be further treated to remove these contaminants. Air stripping and activated carbon absorption are both convenient approaches to remove VOCs [13].

iii) Pre-treatment of raw shale gas FPW. It was reported that coupling MD with appropriate pre-treatment process can remove VOCs and improve the quality of the distillated water product in shale gas FPW treatment [58]. Inspired by this concept, coupling SIE with tailored pre-treatment would allow removal of VOCs before the evaporation step. Suitable pre-treatment options include flotation, coagulation, sedimentation, granular filtration, oxidation, all processes that would reduce the amount of organic contaminants and inorganic scalants from the raw FPW.

5. Potential for valuable resources recovery

5.1. Harvesting of salts

The dominant salt in shale gas FPW is NaCl, and other ions with relatively high abundance include Ca^{2+}, Mg^{2+}, Ba^{2+}, Sr^{2+} [6]. The concentration of these elements in typical shale gas FPW in the U.S. and in China is summarized in Fig. 3b. During the SIE process, these salts will crystallize on the surface of the photothermal materials, providing opportunities for recycling (Fig. 3c) [69]. However, their crystallization will hinder light absorption and decrease the steam generation efficiency. This is a challenge for the long-term and stable operation of the proposed SIE systems in general, especially if the goal is that of achieving ZLD desalination [70]. Recent attempts to solve this problem include preventing salt from adhering to the device as well as increasing salt back diffusion [17, 70]. These methods, however, lead to discontinuous operation or reduced steam generation performance due to the heat loss from fast convection. Moreover, the highly concentrated salt diffuses back into the bulk solution rather than being harvested, wasting the opportunity to obtain valuable mineral resources. Several research works [71-73] overcame this problem by novel designs of localized surface salt precipitation, which is a smart way to keep the system running steadily for a long time and simultaneously harvesting valuable crystalline salts and freshwater. Sodium, magnesium, calcium, barium, and strontium can be recycled as important minerals for industrial activities. For example, recycled NaCl may be used as the raw material for the chlor-alkali industry.

5.2. Recovery of strategic elements

Shale gas FPW is unique in that it often contains non-negligible concentrations of strategic resources [24, 25]. The median values of lithium (Li) concentration in FPW in the U.S. range from 6.6 to 65 mg/L, with Marcellus Shale Region characterized by the highest values. The median value of Li concentration in shale gas FPW in the Sichuan Basin is 33.3 mg/L (Fig. 3b). Although lower than the concentration in salt lakes (>100 mg/L), all values are significantly higher than those in seawater (average 0.17 mg/L [26]), providing great opportunities to harvest Li from shale gas FPW.

Radium (Ra) and Uranium (U) are radioactive elements originated from rocks and crusts. Their concentrations in shale gas FPW are typically low (Fig. 3b) and these elements are difficult to recover due to both complex operation and issues related to risks of radiation. However, they are important elements for nuclear power generation, research, and medicine, thus ranking as strategic resources. U recovery from shale gas FPW may be realized by similar technologies already in place for its harvesting from seawater because of similar concentrations (~3 μg/L) in the two types of stream. Technologies include metal-organic framework membranes, engineered metal-phenolic network membranes, and highly selective adsorbents [74].

Rare earth elements (REEs) play important roles in many high-tech industries, such as aerospace, nuclear, or semiconductor industry. The typical concentration ranges of REEs in shale gas FPW in the Sichuan Basin are reported in Fig. 3d. The total REEs range from 4.5 to 118.3 μg/L, with europium (Eu) characterized by the highest concentrations of 0.92–79.62 μg/L [24]. Eu has wide applications, such as in LED lights, and its market value is expected to reach $0.39 billion by 2026 [24]. The recovery of REEs, especially Eu, from shale gas FPW is of great strategic significance, and also has a bright future.

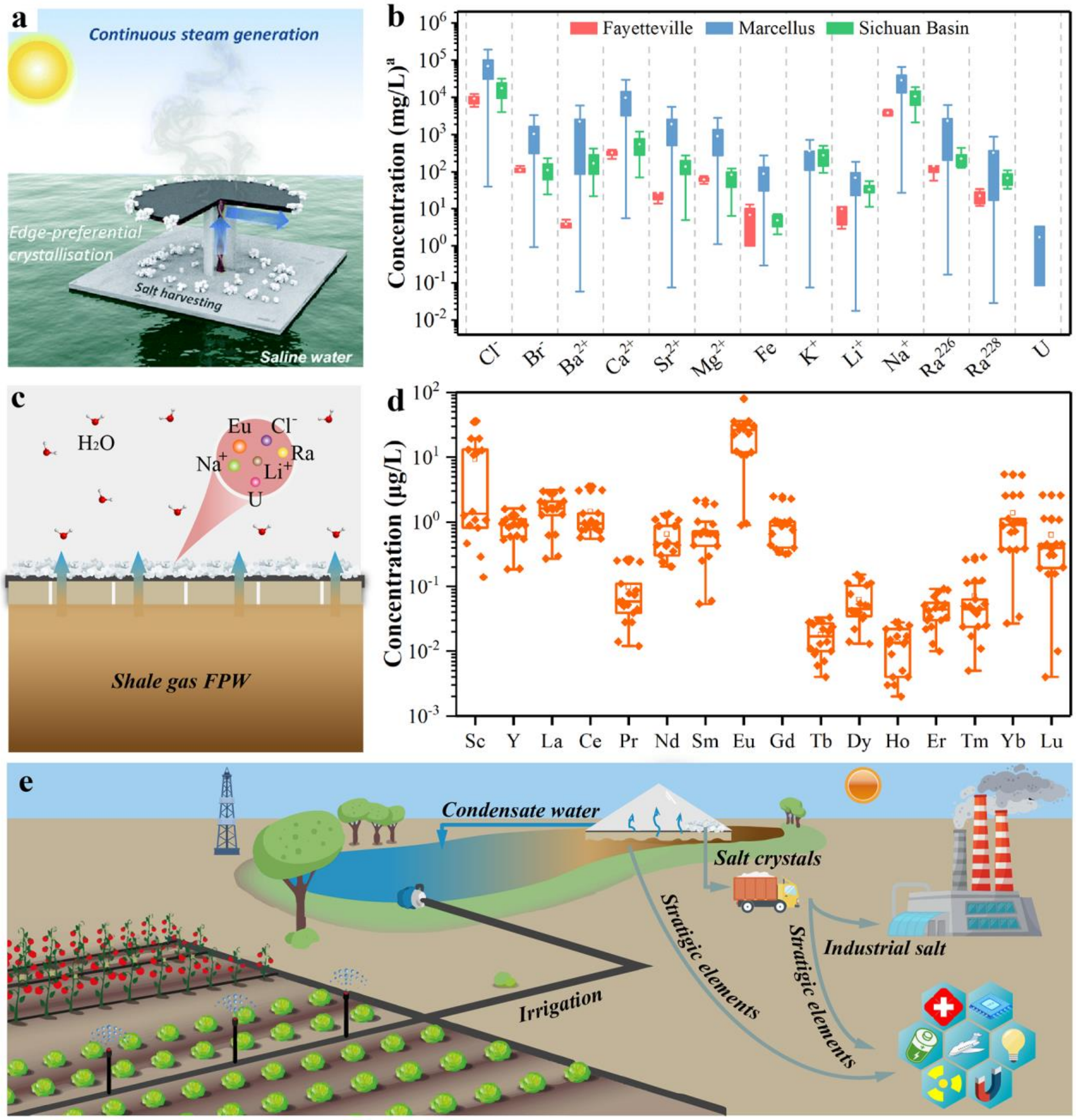

Fig. 3. Valuable resource recovery from shale gas FPW via SIE. (a) Schematic illustration of the novel design for continuous solar steam generation and salt harvesting. Saline water is first transported up to the evaporation disc, and then water evaporates. The remaining salt crystallizes at the edge of the evaporation disc and falls off driven by gravity [71]. (b) Concentrations of most common ions in shale gas FPW from Fayetteville and Marcellus shale plays (the U. S.) [26] and the Sichuan Basin shale plays (China) [11, 75-77]. *Note: The unit of* Ra^{226} *and* Ra^{228} *is pCi/L,*

and the unit of U is μg/L. (c) Schematic of extraction of ions and strategic elements in shale gas FPW from salt crystals. (d) Rare earth elements (REE) concentrations in shale gas FPW from the Sichuan Basin. The raw data refer to the work of Tian et al.[24] (e) Outlook of water reuse for irrigation, salt crystals reuse in industry and strategic elements recovery from raw wastewater and salt crystals.

5.3. Outlook of smart recovery strategies

According to our analysis, solar-driven crystallization is an inexpensive and efficient way to realize resources recovery, however, its drawbacks are also obvious. The crystals are mixed, needing subsequent steps *(e. g.*, selective electrodialysis, permselective exchange membranes in capacitive deionization, supported liquid membrane, ion-imprinted membrane, or ion-sieve membrane) for downstream separation of the different salts, adding cost and complexity to the process. Also, the extraction of low-concentration strategic elements is complicated when the vast majority of the crystals comprise the most abundant and least valuable salts, *i.e.*, NaCl. To tackle this issue, inspired by the principles of ion chromatography, we propose the coupling of solar-driven evaporation device with chromatographic separation to obtain a series of near pure components during crystallization. This technique enables crystallization, separation, and purification in-situ. The separation and purification by traditional chromatography is not perfect and the yield is commonly limited. The development of membrane chromatography may help solving this problem, by achieving continuous flow and high yield. The idea is theoretically possible, but no research has yet combined this process with solar-driven evaporation, which could be a valuable field for future research.

6. Challenges of SIE technology

i) Arguably, the main challenges of treating hypersaline wastewaters, such as FPW, are scaling and fouling phenomena that reduce the long-term efficacy of evaporation [78]. Although a plethora of experimental research efforts are being conducted on SIE to tackle these issues, practical applications are limited. Most of the studies have focused on seawater or lake water, characterized by simpler compositions and lower salinity than FPW. It can be expected that dissolved divalent cations, such as Ca^{2+} and Mg^{2+}, will induce scaling, while dissolved organic matter and bio-species will cause fouling on the solar absorber and within the water delivery structure. Further studies are needed to understand these mechanisms and to find specific solutions to control scaling and fouling. For example, materials preventing salt from adhering to the device should be exploited, and novel designs of localized surface salt precipitation should keep the system running steadily for a long time and simultaneously harvest freshwater and valuable crystalline salts.

ii) The low SWP is an important obstruction limiting the application of SIE. There are several ways to improve the efficiency of SIE in FPW desalination process: improving the solar-thermal conversion efficiency of the solar absorber, reducing heat loss by conduction, radiation, and convection, and improving the thermal efficiency of vapor generation; also, more efficient latent heat recovery. Among them, the most rewarding direction involves the implementation of effective measures for latent heat recovery. A multistage system for latent heat recovery is a key solution to enhance SWP significantly, but no practical application has been implemented so far in SIE technology. Simple, energy-efficient, flexible, lightweight, and compact latent heat recovery strategies should thus be developed for the substantial improvement of the SWP aimed at low-cost SIE systems.

iii) The adaptability of large systems is the third challenge. SIE is suitable for small systems, but its scalability is a difficult task. Efficient large-scale outdoor solar stills should be developed for saline water desalination with inexpensive and commercially available materials.

iv) The salt crystals obtained by SIE process may be classified as hazardous waste and may not meet the standard of industrial salts, because some toxic heavy metal elements, *e.g.*, copper, zinc, chromium (VI), cadmium, may be present and no pretreatment is in place to remove them. This feature will complicate the recovery and reuse of salt crystals, and extra treatment may be required to eliminate hazardous materials and meet purity targets.

7. Outlook

SIE utilizes solar energy as the energy source. The process can be designed by a very simple and inexpensive device, which can be made as an independent integrated system, convenient for transportation and on-site treatment on FPW storage ponds. Furthermore, this technology is not restricted by salinity and can push the desalination process all the way to crystallization. These features make this technique suitable for shale gas FPW treatment, theoretically realizing ZLD.

Potentially, there are two unique additional benefits when using this technology to treat shale gas FPW: water reuse and resources recovery (Fig. 3e), which also contribute to reduce the environmental impacts and meet the principles of circular economy. The condensate water may be reused for irrigation, though a comprehensive evaluation of the water quality is required, and possibly an additional device element or treatment step would be needed to reach the desired quality. Resource recovery, especially of strategic elements, lithium, uranium, europium, REEs, also attracts enormous interest. The smart recovery of salts in the process of water evaporation and salt crystallization, realizing the classified precipitation in one step, is a promising research direction in the near future.

The major challenges of solar-driven evaporation for the beneficial management of FPW are: scaling and fouling; cost-effective latent heat recovery; the scalability of the systems; and efficient water and salts generation. These issues require ad hoc research efforts, analyses, and pilot testing.

Declaration of Competing Interest

The authors declare no competing financial interests.

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