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Reclamation of Produced Water for Beneficial Use

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Abstract: Produced water is the primary waste stream generated during fossil fuel production, with the volumetric water-to-oil ratio approaching 50 : 1 in the later stages of oil field production. In the United States, produced water is usually reinjected, either in support of enhanced oil recovery or for disposal. In arid regions, reusing or recycling the treated water for irrigation or other purposes may be an economically and environmentally attractive alternative.

Successful treatment of complex produced waters generally requires that a series of operations be used to remove different contaminants. Electrodialysis is one possible desalting technique. Recent results indicate that this approach may be appropriate for reclamation of produced waters with relatively low total dissolved solids (TDS) loads but is unlikely to be cost-effective for treatment of concentrated produced waters.

INTRODUCTION

Produced water, also known as formation water or brine, is a by-product of oil and natural gas production. The 17.9 billion barrels of produced water that were generated in 1995 (the most recent year for which data are available) comprised well over 95% of the wastes generated by onshore United States (U.S.) oil and gas exploration and production (E&P) activities (1). The vast majority of produced water is reinjected, either in support of enhanced oil recovery or for disposal.

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A pilot study examined the feasibility of treating oil-field-produced water for industrial, irrigation, and potable use (2, 3), and a variety of possible uses have been proposed for produced water from different sources. The practicality of using produced water for beneficial purposes depends on a number of factors, including the volume of water available, the existence of a local need for water, and the amount of treatment required to meet government or industry-use standards.

Produced water typically contains dissolved salts, with speciation and concentration depending on the water's source. Table 1 uses data for 5 of the 43 provinces in the conterminous United States to illustrate the wide variation in produced water generated with nonassociated gas (4); on average, salt concentrations are similar for oil- and gas-related produced waters (5). Although inorganic ions are the primary contaminants in produced water, volatile or semivolatile organics and free, emulsified, or dissolved oil can also be present.

TECHNOLOGY SELECTION

Site Screening

Geographic information system (GIS) technology is a computer-based tool that can be used to model, analyze, and visualize spatial relationships among data by integrating common database operations with visualization and geographic analysis. With respect to produced-water management, GIS can be used as a first step in locating opportunities for economical reclamation

Table 1. Salt concentrations in produced water generated with nonassociated gas in five provinces (Concentrations in mg/l)

	Appalachian Basin	Denver Basin	Gulf Coast Basin (TX)	Michigan Basin	Permian Basin (TX)
TDS	134,392	7,787	55,479	308,240	112,750
Chloride	81,749	3,839	32,549	193,478	68,880
Sulfate	2,062	103	896	217	1,243
Bicarbonate	569	1,235	889	89	510
Calcium	16,029	72	1,009	37,585	6,407
Magnesium	2,292	34	214	6,737	2,311
Sodium	31,430	2,937	20,331	69,063	29,709

Values are averages extracted from Energy & Environmental Research Center, University of North Dakota; ENSR Consulting and Engineering. *Topical Report. Atlas of Gas-Related Produced Water for 1990*, GRI-95/0016; prepared for the Gas Research Institute and the U.S. Department of Energy, 1995.

and beneficial use. Specifically, GIS can be used to identify areas where supply (significant quantities of produced water) and demand (chronic or acute shortages of water in combination with an existing or anticipated need) intersect. For example, a GIS map integrating precipitation data and U.S. Geological Survey (USGS) produced water data indicates that, while southeastern New Mexico and eastern Wyoming have similarly dry climates, total dissolved solids (TDS) concentrations are significantly lower in Wyoming produced water. Information of this sort can be used to coarsely categorize regions as having greater or lesser potential for reclamation and beneficial use.

Following GIS identification of potential sites, analysis of produced-water samples provides guidance in technology selection. A thorough understanding of the water's composition also allows anticipation of potential equipment fouling or corrosion. While produced water is typically dominated by chloride and sodium (4), a large number of cations and anions may be present at concentrations ranging from 10 ppb to 100 ppm. Table 2 compares inorganic profiles of oil-related produced water and groundwater samples from the Osage–Skiatook Petroleum Research sites near Tulsa. The high sodium, calcium, magnesium, and chloride concentrations shown are typical of produced water in Osage County (6, 7), as are the less dramatic but still elevated strontium, barium, potassium, and iron concentrations (6).

Table 2. Inorganic content of April 2003 water samples from Osage–Skiatook Petroleum Research Sites (concentrations in mg/l)

	Produced water A	Produced water B	Groundwater A
Chloride	103,000	71,600	95.6
Bromide	342	308	0.3
Nitrate	<0.5	<0.5	<0.1
Sulfate	1.6	1.5	5.7
Bicarbonate	63.5	73.2	NA
Calcium	10,700	6,820	50.1
Iron	47.1	24.7	0.2
Potassium	670	130	1.5
Magnesium	1,890	1,610	19.9
Sodium	50,000	38,100	24.0
Lithium	31.6	6.7	<0.5
Barium	468	418	0.4
Manganese	5.7	0.7	1.9
Strontium	474	513	0.3
Chromium	0.02	0.02	<0.02
Ammonium	39.6	57.6	2.9

Notes: Ammonium concentrations are calculated from measured NH_3 values. Bicarbonate concentrations are calculated from measured inorganic carbon values.

Treatment end point is defined by the regulatory and practical standards governing the beneficial use in question. Depending on the application being considered, federal and state regulations may apply; in some cases, multiple agencies have authority. The National Pollution Discharge Elimination System (NPDES) permit program governs point-source discharges to surface soil and surface water, including irrigation (8). Although NPDES is authorized by the federal Clean Water Act, it is typically administered at the state level. The states may also impose additional constraints on beneficial use.

Beyond regulations, there are often practical issues that must be addressed; if reclaimed produced water is to be used for irrigation, salinity (typically represented by electrical conductivity) and sodicity are of particular concern. High salinity reduces the water available in the soil matrix, limits root absorption, and, in turn, leads to stunted crop growth and loss of yield (9–11). The salinity threshold at which yield begins to suffer is crop dependent (10).

Sodium adsorption ratio (SAR) is another critical parameter and is defined in Eq. (1):

$$\mathrm{SAR} = \frac{[\mathrm{Na}^+]}{\sqrt{\left([\mathrm{Ca}^{2+}] + [\mathrm{Mg}^{2+}]\right)/2}}, \text{ concentrations in meq/l} \qquad (1)$$

High SAR irrigation water can adversely affect soil permeability and water penetration. Advisable SAR values are crop and soil dependent and are also affected by salinity, but waters with conductivity below 1.5 dS/m and SAR below 6 can generally be used safely (11). Salinity and sodicity are especially likely to be problematic in arid regions, where salt accumulated in soil is less likely to be diluted by precipitation than it would be in wetter areas. For this reason, these parameters are of particular importance in the regions where reclaimed produced water is most likely to be used for irrigation and other ground applications.

While these constraints are specific to irrigation, analogous sets of requirements apply to other potential beneficial uses [e.g., the United Nations document that discusses irrigation also provides guidance on water quality standards for livestock and poultry watering (9)]. Obviously, water intended for human ingestion is required to meet exceptionally high standards.

Laboratory Evaluation

After the considerations described above have been used to define a specific treatment scenario, candidate technologies can be selected for evaluation. Table 3 lists methods that have been studied at bench, pilot, and demonstration scales. In many cases, multiple processes are needed to remove the different contaminants in a given produced water (2, 3, 12–15).

Table 3. Examples of potential produced-water treatment technologies

Technologies	References
Organics removal	
Adsorption	16
Biological treatment	12, 13
Induced air flotation	2, 3, 13
Membrane-based separations	
Ultrafiltration	17, 18, 19
Microfiltration	18, 20
Inorganics removal	
Freeze/thaw evaporation	21
Ion exchange	2, 3, 14
Membrane-based separations	
Electrodialysis	13, 15
Microfiltration	20
Nanofiltration	17, 22
Reverse osmosis	14, 15, 17, 22
Precipitation	2, 3
Combined	
Constructed wetlands	23

Given the predominance of inorganic over organic contaminants and the importance of conductivity and SAR, effective desalting is critical. Recent studies focused on electrodialysis (ED), a membrane-based technology that uses an electric field to drive migration of ionic species from a diluate or feed solution into a more concentrated brine. The ED stack is composed of alternating anion and cation exchange membranes, which are permeable to negatively and positively charged ions, respectively. Under influence of an applied electric field, cations migrate toward the cathode at one end of the stack, while anions travel toward the anion at the opposite end. The charge selectivity of the ion exchange membranes results in accumulation of ions in alternating compartments within the stack; these concentrate cells alternate with diluate cells, which are ion-depleted. A schematic diagram appears in Fig. 1. Detailed information on ED theory, apparatus, and applications is provided by Strathmann (24). In addition to concentrating seawater for production of table salt (24), ED has been used to reduce TDS concentrations in a variety of waters, including surface water (25), groundwater (26), and brackish water (24). The feasibility of using ED for produced-water treatment has also been considered (13, 15).

During the summer of 2003, the Gas Technology Institute (GTI) conducted a series of tests with the goal of evaluating the ability of ED to reduce TDS concentrations in different produced waters. An Ameridia EUR2B-10 skid with 10 cell pairs was used. Each cell pair included a Neosepta® CMX-SB cation-exchange

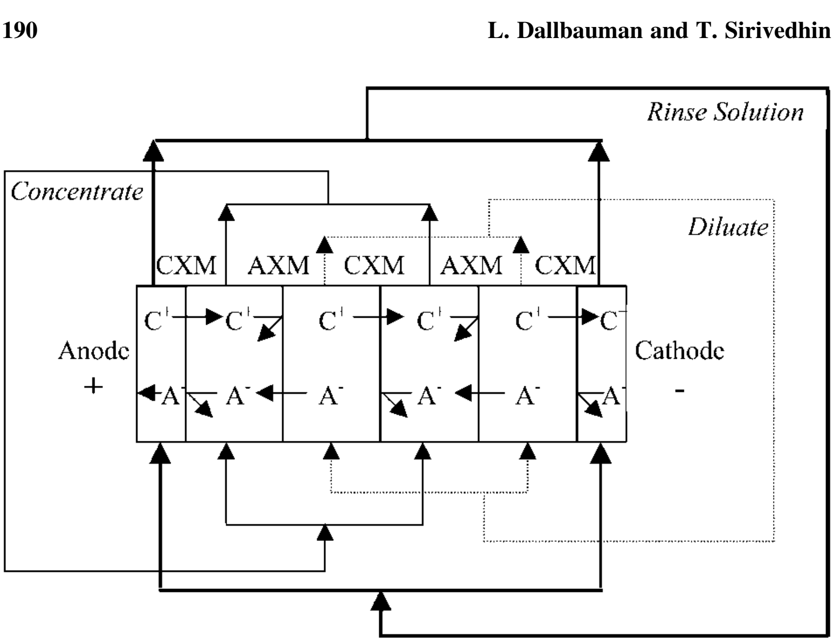

Figure 1. Schematic representation of electrodialysis stack (CXM = cation exchange membrane; AXM = anion exchange membrane; C^+ = cation; A^- = anion).

membrane and a Neosepta® AMX-SB anion-exchange membrane. In addition, two CMX-SB membranes were used as end membranes to prevent contact between the diluate/concentrate and the electrode rinse solutions. The total surface area of each cell was 2000 cm^2; active area was 200 cm^2.

Diluate and concentrate flow rates were maintained at 3.5 L/minute (0.92 gpm) and the electrode rinse solution (3 M $NaNO_3$) flow rate at 3.8 L/minute (1.0 gpm) to each electrode compartment. The system was operated in batch mode, with each experiment starting with 4 L of feed and 4 L of 25 g/L NaCl, which ultimately became the concentrate stream. The stack was operated in constant voltage mode, with settings of 3.0, 6.5, and 9.8 V.

A ThermoOrion 105A Plus conductivity meter and a Markson pH-Vision 6071 were used to measure conductivity and pH directly in the diluate compartment. A total of 50 mL (10 samples of 5 mL each) was drawn from the diluate compartment with a glass pipette. A Perkin Elmer 4300 DV optical emission spectrometer was used to measure cation concentrations. A Dionex 2010i ion chromatograph was used to determine anion concentrations. A Rosemount Analytical Dohrmann DC-190 was used to measure bicarbonate by combustion-infrared detection, and the phenate method [4500-NH_3 F; (27)] was used to measure ammonia.

Chemicals used to prepare test solutions included $CaCl_2 \cdot 2H_2O$, LiCl, $MgCl_2 \cdot 2H_2O$, $NaHCO_3$, NaBr, Na_2SO_4, (all Fisher Scientific, ACS grade), NaCl (Fisher Scientific, biological grade), KCl (Mallinckrodt Chemical),

Table 4. Low-salt produced-water recipes

		Concentration (mg/l)							
State	pH	K^+	Na^+	Ca^{2+}	Mg^{2+}	Cl^-	HCO_3^-	SO_4^{2-}	TDS
CO	8.2	29	1,711	10	1	1,000	2,196	555	5,502
TX	7.5	9	1,606	1	1	2,024	763	30	4,435
WY	8.2	5	1,538	10	4	130	1,122	2,200	5,009

NH_4Cl (Sigma, ACS grade), and HCl (Fisher Scientific). Deionized water was used for the preparation of all solutions.

Results: Low-Salt Produced Waters

Review of the USGS database (7) allowed identification of produced waters with similar TDS concentrations (~5000 ppm) but different speciation. Three of these waters were selected for testing. They were from different basins and represent a range of monovalent (Cl^- and HCO_3^-) and divalent (SO_4^{2-}) anion concentrations, as indicated in Table 4. Due to concerns about potential

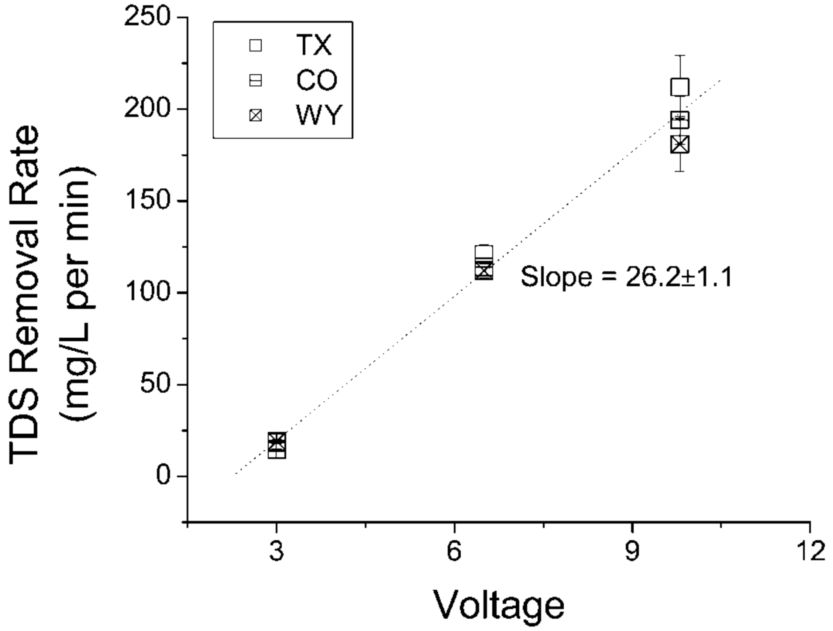

Figure 2. Rate of TDS reduction as function of voltage for CO, TX, and WY recipes.

membrane fouling (28), candidate waters for this test series were limited to those having calcium and magnesium concentrations under 10 mg/L.

The influence of applied voltage on TDS reduction was studied. Figure 2 shows the rate of reduction at each voltage setting for the three feed streams. Within experimental and analytical errors, the reduction rates for the three streams were equal and increased linearly with voltage, with reduction rate increasing by 26.2 mg/L per minute for each unit increase in voltage.

The decrease in SO_4^{2-} and HCO_3^- concentrations with time is illustrated by Fig. 3; trends for Cl^- were similar to those for HCO_3^-. The concentration of each species is normalized with respect to its own initial concentration so that the plots can be used to compare fractional removal over time. Figure 3A shows that at low voltage, bicarbonate removal (i.e., slope) was greatest for the recipe representing the CO produced water, which had the highest concentration of that ion and thus also the highest absolute removal rate. As voltage increased, removal rate increased for all three recipes, and fractional removal of bicarbonate was approximately equal for all three at 9.8 V, as illustrated by Fig. 3C. In contrast, comparison of Figs. 3B and 3D shows that at low voltage, fractional sulfate removal was lowest for the recipe with the highest sulfate concentration (WY) and that removal rate again increased with increasing voltages for all three recipes until fractional removal was approximately equal at 9.8 V.

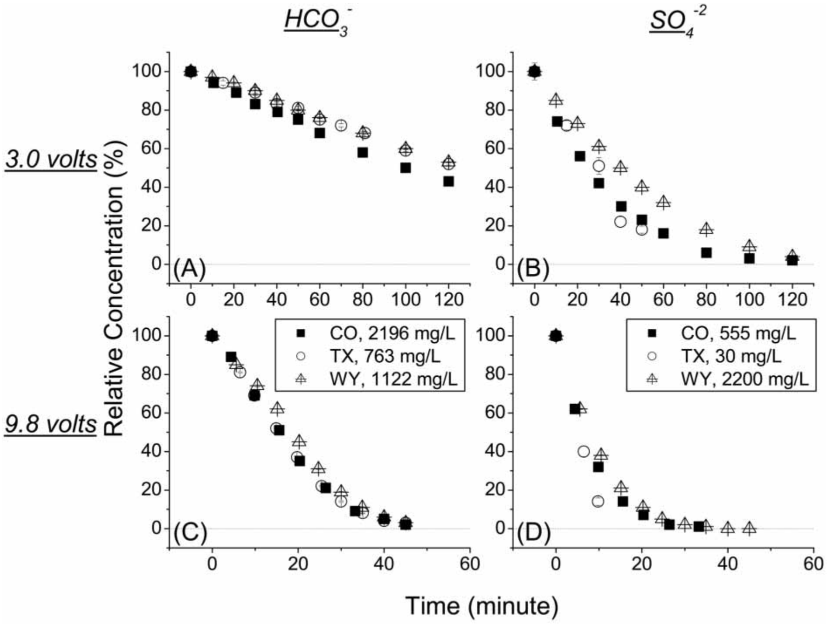

Figure 3. Decrease in concentrations of (A) HCO_3^- at 3.0 volts, (B) SO_4^{2-} at 3.0 volts, (C) HCO_3^- at 9.8 volts, and (D) SO_4^{2-} at 9.8 volts for CO, TX, and WY recipes.

Table 5 summarizes the initial and final conductivity and SAR values for the three low-salt recipes. The final conductivities of all three were well below the "generally safe" conductivity of 1.5 dS/m for applied ED voltages of 6.5 and 9.8 V but are greater than 4 dS/m at 3.0 V ED potential. This fact, combined with the relatively modest reduction in ionic concentrations that were achieved after 2 h at 3.0 V, indicates that ED at this low voltage is impractical even for these relatively clean produced waters.

Because these recipes were deliberately based on produced waters with low divalent cation concentrations, their initial SAR values were high. At low voltage, final SAR values were even higher because the removal of a relatively small number of sodium ions produced a small decrease in the numerator of Eq. (1), while removal of a small number of calcium and magnesium ions resulted in a large decrease in the denominator. As voltage increased, a larger number of sodium ions was removed, but there were relatively few calcium and magnesium ions available; hence, the denominator remained fairly constant while the numerator decreased, allowing SAR to decrease. In practice, gypsum or other amendments have been used to

Table 5. Initial and final conductivity and SAR for low-salt produced-water recipes

	CO		TX		WY	
	Initial	Final	Initial	Final	Initial	Final
3.0 V: Time, min		120		120		120
Conductivity, dS/m	7.13	4.87	7.66	4.72	6.36	4.43
Na^+, mg/L	1,760	1,020	1,580	939	1,560	963
Ca^{2+}, mg/L	5.33	0.19	0.76	0.11	6.18	0.10
Mg^{2+}, mg/L	0.62	<0.05	0.69	<0.05	2.52	<0.05
SAR	192	>538	316	>589	134	>621
6.5 V: Time, min		60.2		60		60
Conductivity, dS/m	7.16	0.50	7.49	0.27	6.50	0.54
Na^+, mg/L	1,660	110	1,520	52.2	1,620	108
Ca^{2+}, mg/L	6.17	1.03	0.61	0.17	7.25	0.58
Mg^{2+}, mg/L	0.71	<0.05	0.50	<0.05	2.83	<0.05
SAR	169	>28.7	349	>28.2	124	>36.5
9.8 V: Time, min		50		45		45
Conductivity, dS/m	7.18	0.08	7.83	0.06	6.35	0.09
Na^+, mg/L	1,740	20.7	1,610	13.9	1,540	21.0
Ca^{2+}, mg/L	5.0	0.64	0.58	<0.05	6.94	0.11
Mg^{2+}, mg/L	0.59	<0.05	0.55	<0.05	2.36	<0.05
SAR	196	6.7	364	>10.5	129	>13.2

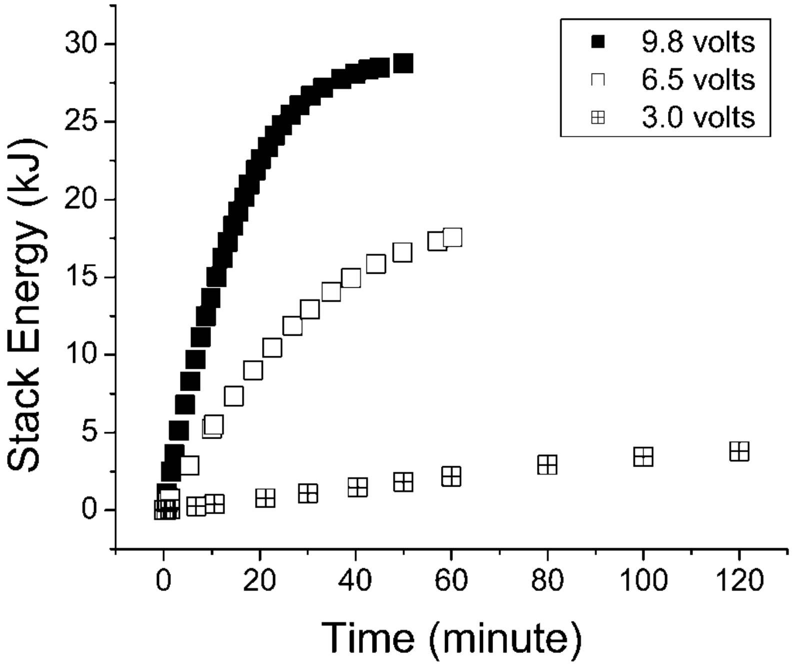

Figure 4. Cumulative stack energy usage at 3.0, 6.5, and 9.8 volts for CO recipe.

increase calcium concentration (and decrease SAR values) of coal-bed methane produced water before it is used for irrigation (8, 29, 30).

As stack energy consumption is likely to be the main contributor to ED operating costs, this value was calculated and is shown in Fig. 4 for the CO recipe at 3.0, 6.5, and 9.8 V. Results are similar for the TX and WY recipes. As expected, the advantage of greater reductions in conductivity (and thus ionic concentrations) that could be achieved in shorter times at higher voltages was offset by an increase in power consumption. The relative importance of processing time, energy costs, and water quality must be determined on a case-by-case basis.

Results: High-Salt Produced Water

In addition to the low-salt produced waters just described, the ability of ED to treat high-salt produced water was evaluated. A recipe based on the produced water B sample from the Osage–Skiatook Petroleum Research sites (see Table 2) was developed. The recipe had significantly lower TDS than the sample. Because of concerns about possible fouling of the ED membranes, divalent cations were omitted altogether, and electroneutrality required that the elimination of the high concentrations of Ca^{2+} and Mg^{2+} be balanced by

Table 6. Monovalent Osage-Skiatook recipe

	Monovalent recipe	Produced water B
Chloride	58,864	71,600
Bromide	308	308
Nitrate	0	<0.5
Sulfate	1	1.5
Bicarbonate	73	73.2
Calcium	0	6,820
Iron	0	24.7
Potassium	130	130
Magnesium	0	1,610
Sodium	38,114	38,100
Lithium	7	6.7
Barium	0	418
Manganese	0	0.7
Strontium	0	513
Chromium	0	0.02
Ammonium	58	57.6

a reduction in the concentration of the dominant anion (Cl^-). The recipe, which is listed in Table 6, could represent a produced water that has undergone a pretreatment stage, such as precipitation to remove calcium and magnesium salts.

The tests performed on this water were similar to those described above; but, as expected, the high TDS concentration led to significantly different results. TDS reduction rate was again linear with voltage but increased at a greater rate, as illustrated by Fig. 5. Figure 6 compares the decrease in feed conductivity for the CO and OK recipes. These data were obtained at 9.8 V. While the conductivity of the CO water falls within the satisfactory range (5–8 dS/m) for livestock watering (9) before it has been treated, the conductivity of the OK water exceeds the 16 dS/m maximum for "very limited" livestock watering (9) after more than 2 h of processing. The energy used to achieve these results is plotted in Fig. 7, which, in combination with Fig. 6, illustrates the challenge facing reclamation of concentrated produced water. After expending over 500 kJ in 140 min (more than 60 W), the 4 L of OK water processed fail to meet minimal standards for irrigation or livestock watering, two of the most likely applications for reclaimed produced water.

Postscript: Effects of Divalent Cations on ED Performance

Following completion of high and low salt testing, a series of tests aimed at evaluating the likelihood of Ca^{2+} to foul the ED membranes was initiated. Mass balance calculations indicate that no calcium is accumulated in the

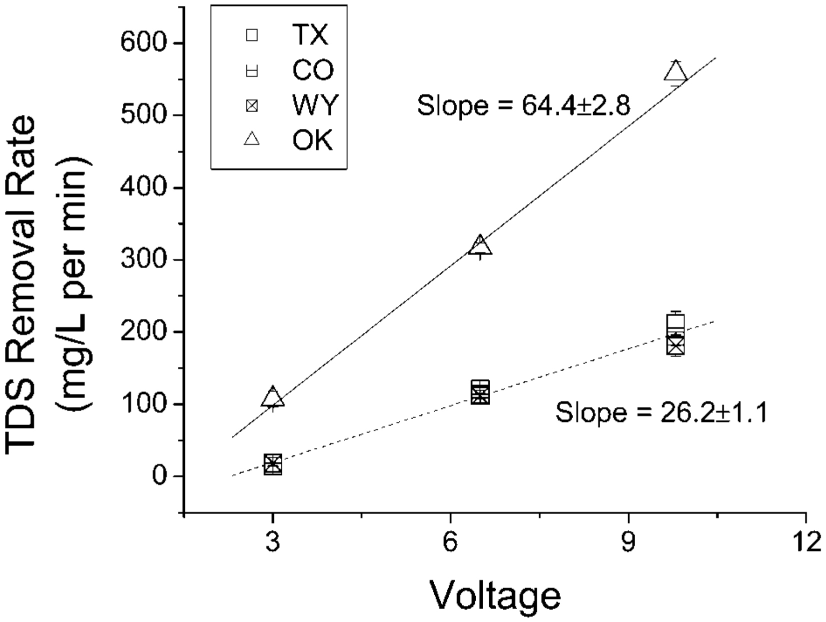

Figure 5. Comparison of TDS reduction rate as function of voltage for high- and low-salt recipes (solid line = high-salt recipes; dotted line = low-salt recipes).

stack for feed concentrations up to 100 mg/L, and no performance degradation was observed for concentrations up to 150 mg/L. These results suggest that the ED stack can be used to process more realistic produced-water recipes, although the exceptionally high calcium concentrations in the Osage–Skiatook produced water remain problematic.

SUMMARY

The produced water generated with oil and natural gas has historically been viewed as a liability. In light of ever-increasing demands for water, the possibility of turning produced water into an asset via reclamation for beneficial use is receiving increased attention. In order for this conversion to succeed, locally appropriate reclamation approaches must be defined. After sources of produced water have been identified and matched with real-life beneficial uses, produced-water composition, water-quality requirements, and local concerns define candidate technologies for evaluation.

Electrodialysis is one possible approach to desalting produced water. The ability of ED to treat low- and high-salt produced waters was studied at laboratory scale. Although energy costs are likely to preclude using ED to treat

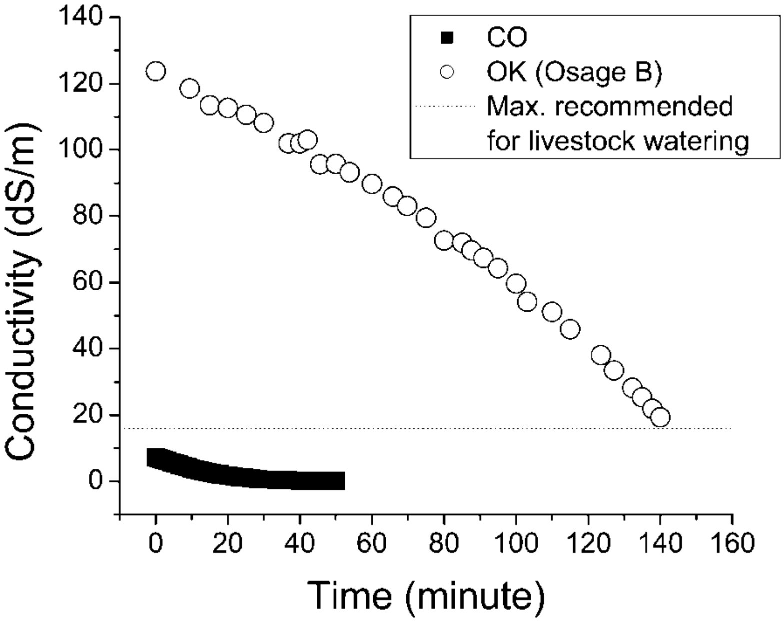

Figure 6. Decrease in conductivity at 9.8 volts for OK and CO recipes.

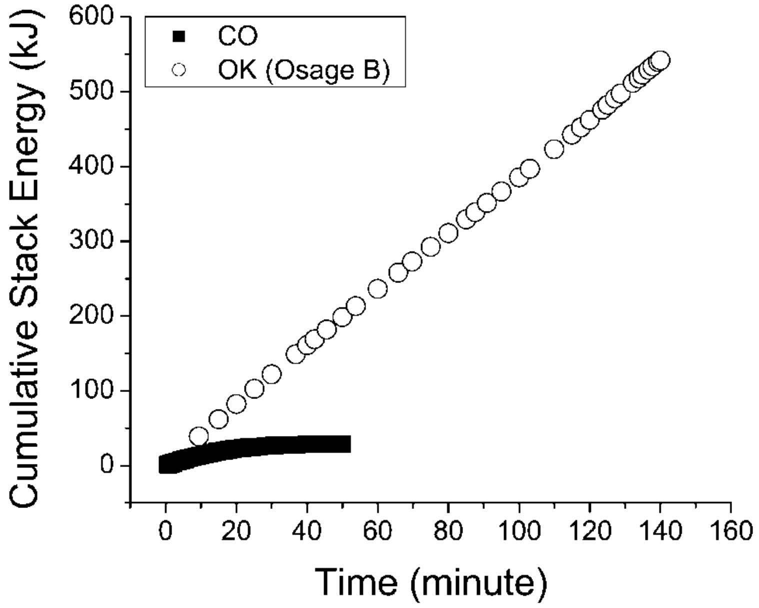

Figure 7. Cumulative stack energy usage at 9.8 volts for OK and CO recipes.

concentrated-produced water, the technology shows promise for treatment of relatively clean produced waters.

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