Optimal Design of Thermal Membrane Distillation Systems for the Treatment of Shale Gas Flowback Water

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Abstract: Shale gas production is associated with the significant consumption of fresh water and discharge of wastewater. The flowback wastewater is tied to the hydraulic fracturing technology used for completing and stimulating the horizontal wells in the very tight formations characterizing the shale formation. Treatment and reuse of these large volumes of wastewater can lead to substantial savings in fresh water usage and reduction of the negative environmental impact thereby enhancing sustainability of the shale gas industry. Such treatment requires selective and cost-effective technology.

Thermal membrane distillation (TMD) is an emerging technology that offers several advatanges such as high selectivity in separating water from inorganic solutes and modular nature that can accommodate a wide range of flows. It can also utilize low-level heats that are typically available from shale-gas production and processing.

The objective of this work is to develop an optimization approach for the design of TMD systems to treat flowback water. A multi-period formulation is developed to account for the time-based variation in the flowrate and concentration of the flowback water. Modeling equations are used to relate design and operating variables to performance and cost. The optimization formulation also accounts for the period-based changes in the required design and operating variables and reconciles them over the selected periods. Other constraints include quality of the permeate and water-recovery ratio. The optimization formulation and design approach are applied to a case study for the treatment of flowback water for the Marcellus Shale Play. For 75% water recovery, the cost of the permeate is about \$2.6/m^3. As higher recoveries are sought, the cost per m^3 of permeate increases due to capital productivity factors in dealing with the decreasing amount of flowback water over time. The results are reported using a Pareto chart that trades off recovery objectives with cost of treated water.

Keywords: Thermal membrane distillation, Shale gas production, Flowback water, Process integration, Scheduling optimization.

INTRODUCTION

The US is witnessing a substantial growth in the production of shale gas. It is expected to become one of the most important energy sources in the US. Shale is a sedimentary rock characterized by its very tight formation compared with the conventional gas basins. Therefore, it requires special techniques for well completion including the drilling horizontal wells and the use of hydraulic fracturing for production stimulation. It is estimated that over 35,000 wells are hydraulically fractured annually in the largest five reserves in the US are the Barnett, Fayetteville, Haynesville, Marcellus, and Woodford plays [1].

Water is considered one of the most important and limiting commodities associated with shale gas production. Fresh water is heavily consumed in the pre-production stages including site preparation, drilling, and completing the well. Hydraulic fracturing (fracturing) technology is a stimulation technique used for shale gas wells. It involves the injection of large volumes of water-based fracturing fluid through the tight formation to create artificial pathways through which gas and oil may be produced. The volume of water needed for fracturing a well varies based on several factors including depth of the well, length and numbers of planned fracks, and the geology of the play formation [2]. In some cases, water needed for drilling and fracturing was estimated to lie between 2-6 million gallons of water per well with about 89% percent of this amount used in well fracturing [1]. Fracturing fluids are typically mixtures of water and chemicals with water forming the bulk (*e.g*., 98%) of the fracturing fluid volume [1]. Generally the main fracturing additives for the fracturing fluids are: sand, friction reducers, gelling agent, acids, surfactants and proppants.

The typical resources of the substantial volumes of water needed during shale gas fracturing are surface

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fresh water bodies and underground water. With the increasing demand for water resources and the shortage of water supply in several shale-gas production areas, there is a critical need to treat and reuse wastewater discharges from shale production. This approach also reduces the negative impact on the environment. Wastewater is discharged in large volumes during shale gas production. Flowback wastewater is generated during the well completion stages while produced water is generated during the production stages. Flowback water is typically recovered within the first 30 days after injecting the fracturing fluids. Typically, 10-40% of the total injected water and the rest of the injected fluids remain in the formation [3]). For instance, in the Marcellus Shale Play, about 10-15% of the fracturing fluid is reproduced as flowback representing about 600,000 gallon per well in the first 10 days of the fracturing process [1]. The selection of the management option to handle the large volumes of the flowback water involves several factors such as: economics, environmental regulations, volumes and composition of the flowback water, and availability of fresh water sources.

The current management options for the flowback water vary significantly and each shale play has its own management strategy. Management options include:

Disposal

Disposing the flowback water is considered a plausible option if certain criteria are met:

- Availability of discharge outlets
- Acceptability according to environmental regulations
- Sufficiency in the supply of fresh or underground water resources
- Safety of transportation to disposal outlets

Disposal includes three primary options: underground injection, surface discharge, and pond evaporation. There are several environmental regulations governing disposal of flowback water. There are also potential public health and safety concerns. Finally, disposal does not promote water-reuse strategies. It merely enables the discharge of wastewater.

Treatment

This alternative renders the flowback wastewater in a condition that is acceptable for discharge or even reuse in other fracturing jobs. The selection of a treatment options depends on the volume of flowback, total dissolved solid (TDS) levels, and the relative performance compared to using fresh water and injecting in wells. When the volumes of flowback are huge, the underground injection option is either unavailable, unsafe or uneconomical. Different treatment levels may applied to the flowback water enabling further reuse. Typically, the first level of treatment “pretreatment” consists of removing the organics and total suspended solids (TSS). The flowback water is subjected to filters, with pore sizes ranges from 0.04-3.00 microns, to remove TSS and sand [1]. After pretreatment, flowback water containing low to moderate levels of TDS may be blended with fresh water and additives then reintroduced as a fracturing fluid. For high TDS concentrations, flowback water may require further treatment to reach the reuse targets. Typically, the advanced levels of treatment include desalination to treat the high levels of TDS. Typically, the content of TDS in the flowback water ranges from around 40,000 mg/L in the Barnett Shale Play to 150,000 mg/L in the Bakken Shale Play [3]. It may even reach 280,000 mg/L in some of the high-salinity flowback water streams in the Marcellus Shale Play. Highly saline flowback requires substantial desalination to enable recycle, reuse or even discharge. Such desalination is needed for various reasons including the following:

- Reuse in fracturing fluids requires low salinity to avoid formation damage and destruction of drilling equipment [2].
- High levels of salinity can negatively interfere with the performance of some friction reducers required in the fracturing fluid [2].
- The quality of the flowback deteriorates progressively over the fracturing period which makes it difficult to directly recycle the water or dilute without being treatment. This phenomenon occurs because of increasing the contact time between the fracturing water and the shale rock and formation water will lead to elevated levels of contaminant in the returned flowback [3].

The choice of the desalination technology is subject to: the desired level of treatment, economics, mobility, and adjustable configuration of the treatment units. The most common contaminants found in flowback water are residual chemicals from additives injected with the fracturing fluid including (biocides, scaling inhibitor, and friction reducers) as well as salts, metals and organic compounds originally found in the formation and formations water which returns back within the flowback. The concentration of the flowback water may vary significantly depending on location and formation characteristics. Another important factor in treating flowback is the fluctuation in the rate of collected water. Typically the volume of the collected flowback water is initially high then it decreases dramatically until it diminishes when the production phase starts and the produced water is generated [3]. Therefore, there is a critical need for cost-effective treatment technologies that are highly selective and adaptable to varying flowrates and compositions of the flowback water.

Thermal membrane distillation (TMD) is a highly selective membrane technology that can be used effectively to treat wastewater from gas production facilities. The feed is preheated to a temperature below boiling and is introduced into a shell that contains a selective hydrophobic membrane. The water vapor passes through the membrane and is condensed on the other side using a sweeping stream (typically the stored permeate). In addition to heat transfer from the heating agent to the feed, there is also heat transfer across the membrane which is coupled with the mass transfer of the water vapor (convective heat transfer) as well as conductive heat transfer across the membrane material. The sweeping water is used in large enough quantities to condense the permeating vapor and to prevent buildup in heat at the membrane surface. Figure **1** is a schematic representation of a direct contact TMD system.

The use of TMD offers several attractive features for flowback desalination including [4, 5]:

- Mobility and Compactness: the TMD modules have a small footprint because of the large surface area to volume ratio of the membrane
- Very high rejection of salts allowing obtaining the desired permeate quality
- Ability to handle high concentrations of salts generally found in flowback
- Modularity with relatively easy addition and removal of TMD modules to adjust to the required capacities
- The TMD modules can also be skid mounted and moved from application to the other. This aspect is important in shale gas production because of the fracturing of various well during the exploration phase.
- Usage of low-level heat to effect the separation. This is a particularly relevant factor in shale gas production because of the availability of several sources for inexpensive low-level heating such as flaring. After a fracturing job is completed, flaring is initiated for well testing purposes. Flaring is a huge source of wasted energy that may be utilized for providing a free source of heating to fulfil the TMD energy requirement.

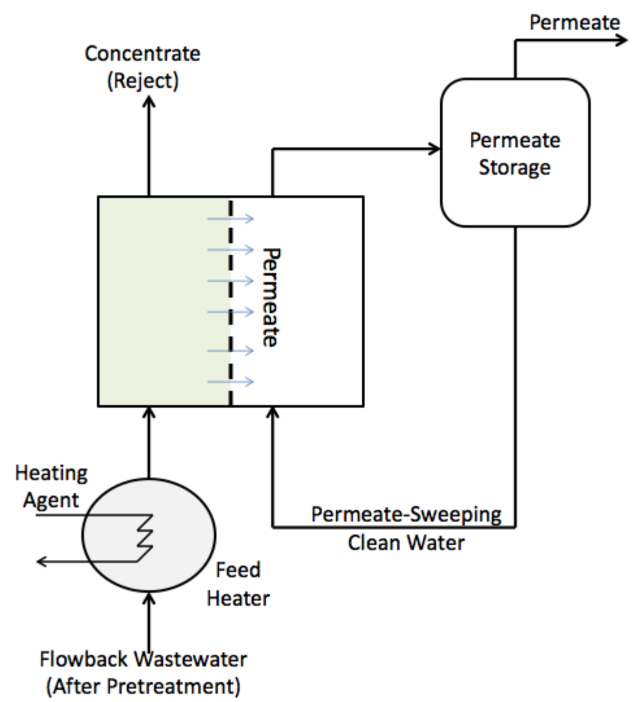

Figure 1: A Schematic Representation of Direct Contact TMD System.

The design studies of TMD [6-9] have been based on a feed with constant characteristics (flowrate and composition). In flowback applications, there is a continuous change of flowback flowrate and composition. The objective of this work is to introduce a new approach for the design of TMD systems with varying feed characteristics and to apply it to the treatment of flowback wastewater. The design approach employs a multi-period optimization formulation to handle the variations in flowrate and concentration of the flowback and to minimize the overall cost of the system while reconciling operating and fixed costs. The approach is also used to tradeoff extent of treatment (recovery) and cost. A case study for the Marcellus Shale Play is used to demonstrate the merits and applicability of the devised approach.

PROBLEM STATEMENT

Consider a shale play with planned fracking activities for multiple wells over a certain decision-making time horizon. As a result, flowback wastewater will be generated. A mobile TMD system is to be designed to treat the flowback water and to be moved from one well to the other depending on demand. The generated flowback wastewater streams have known (expected) flow rate and composition profiles over the decision-making time horizon. The flowback wastewater is first pretreated to remove oils and organics. The inorganic dissolved salts are to be removed using TMD. The pretreated flowback water is to be heated to a certain temperature, T_{FB}, then fed to TMD. The quality of the treated water (permeate) must meet specific quality requirement corresponding to reuse or discharge. The reject must also satisfy certain constraints on composition (*e.g.*, to avoid precipitation or to allow disposal). A minimum amount of the treated water is to be collected. Available for service is a certain rate of heating energy source (Q_{flare}) from the flaring that is associated with the well testing. Available for service is also a set of external (purchased) heating utilities to be used as needed supply additional energy for TMD. A network of mobile TMD modules is to be used for desalinating pretreated FB. The total area, number of modules, design specifications, and operating conditions for the TMD network are to be determined so as to minimize the cost of the system. Figure **2** is a schematic representation to the problem statement.

APPROACH

In order to streamline the optimization formulation, a multi-period approach is developed based on the following considerations:

- The TMD system is used to treat the flowback water from one well then it is moved to the next well and so on. For each well, there is a cycle time, τ_c , over which the flowback water is treated.
- The decision-making time horizon for the design of the TMD system is the summation of all consecutive cycle times, *i.e*.

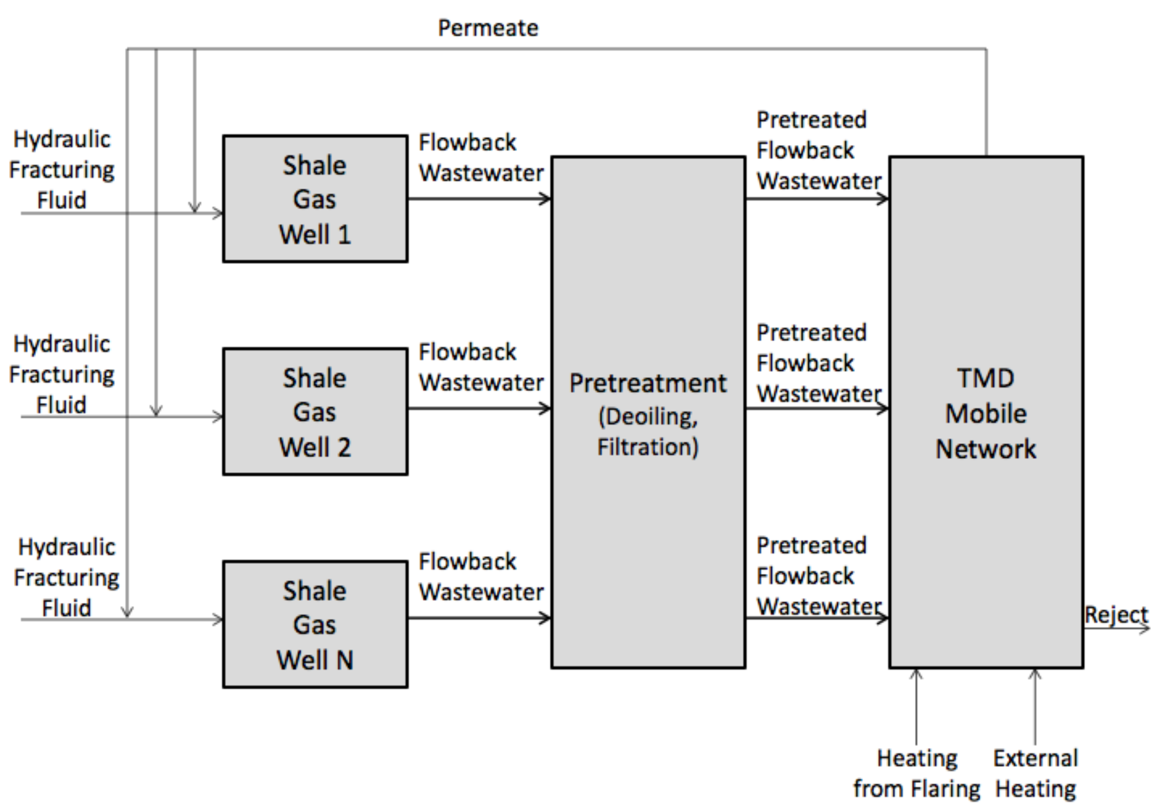

Figure 2: Representation of the problem statement.

Decision-making time horizon = $\sum_c \tau_c$ (1)

- The decision-making time horizon (*e.g.*, a year) is discretized into N_t periods (*e.g.*, each period corresponding to one day). The index t is used to characterize the periods and the set of periods within the decision-making time horizon is described by PERIODS = {t|t= 1, 2, …,$N_{Periods}$}. The key characteristic of a period is that average flowback characteristics (flowrate $W_{FB,t}$ and composition $y_{FB,t}$) are taken to be constant during that period. Figures **3a** and **3b** illustrate this discretization. First, Figure **3a** shows the continuous change in flowback flowrate over time for the multiple cycles. Then, Figure **2b** demonstrate the discretization over the multiple periods. The multi-period approach enables tackling of the dynamic system through a sequence of steady-state models each corresponding to a time period. For the t[th] period, the vector of TMD modelling equations is given by:

$$\Phi_t(W_{FB,t}, y_{FB,t}, D_t, O_t, S_t) = 0 \qquad \forall t \qquad (2)$$

where D_t, O_t, and S_t are the vectors for the design, operating, and state variables, respectively.

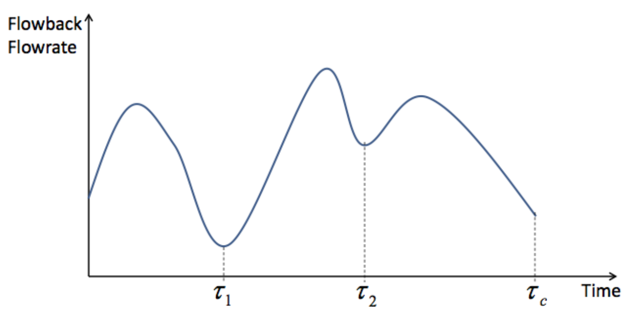

Figure 3a: Flowrate Fluctuation over Cycles.

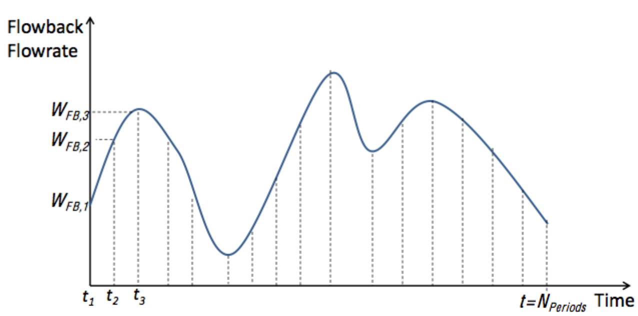

Figure 3b: Multi-Period Discretization of Flowrate.

The objective function is aimed at minimizing the total annualized cost of the TMD water-management system:

Minimize total annualized cost = AFC + $\sum_{t=1}^{N_{Periods}} Op\_Cost_t * H_t$ (3)

where AFC is the annualized fixed cost of the system, Op_Cost_t is the operating cost per hour, and H_t is the number of operating hours in period t.

The objective function is subject to the following constraints:

Modeling equations:

$$\Phi_t(W_{FB,t}, y_{FB,t}, D_t, O_t, S_t) = 0 \qquad \forall t \qquad (4)$$

It is worth noting that the system size as represented by the design vector D_t (*e.g.*, membrane area) may vary over the operating periods. When the actual system is constructed, it has to be sized to be large enough to handle all the required tasks for all periods including the largest required design vector (which is to be determined through optimization). Therefore, the design vector to be used, $\bar{D}$, should satisfy the following constraint:

$$\bar{D} = \arg\max\{D_t \mid t = 1,2,...,N_{Periods}\} \qquad (5)$$

Alternatively,

$$D_t \le \bar{D} \qquad \forall t \qquad (6)$$

The annualized fixed cost of the TMD system is a function of $\bar{D}$ through a cost function:

$$AFC = \Omega(\bar{D}) \qquad (7)$$

Additional constraints include:

Quality constraints of the collected permeate:

$$y_{Permeate,t} \le y_{Desired,t} \qquad \forall t \qquad (8)$$

Concentration limits on the reject composition to avoid precipitation and scaling:

$$y_{\mathrm{Reject,t}} \le y_{\mathrm{limit,t}} \qquad \forall t \qquad (9)$$

Minimum recovery requirement:

$$\sum_{t=1}^{N_{Periods}} W_{Permeate,t} * H_t \geq Permeate_{Desired} \tag{10}$$

where $W_{Permeate,t}$ is the flowrate on an hourly basis of the collected permeate during period t, H_t is the number of hours during period t, and $Permeate_{Desired}$ is the total desired amount of permeate to be collected over the decision-making time horizon.

CASE STUDY

The data for the flowback water in this case study are taken from information provided by Hayes (2009) for Location E which corresponds to a horizontal well in the Marcellus Shale. The data collected on days 1, 5, and 14 from the hydraulic fracturing event are reported by Table **1.** The bulk of the recovered flowback water is recovered over the first 14 days. Tracking of the rest of the flowback water up to 90 days confirms this observation [10].

Table 1: Flowback Water Data for the Case Study [10]

Day	Cumulative Volume of Collected Flowback Water, BBL	TDS in Collected Flowback Water, ppmw
1	8,560	28,900
5	20,330	55,100
14	24610	124,000

The flowback data were correlated using regression technique to obtain the following functions:

$$\text{Cumulative flowback water (BBL)} = 6{,}190 * \ln(t) + 9{,}067.3 \tag{11}$$

and

TDS in collected flowback water (ppmw) = 7,372.9*t + 20,180 $R^2 = 0.999$ (12)

where t is in days from the hydraulic fracturing event and t lies between 1 and 14 days. Next, the cumulative flowback correlation was used to determine the collected volume each day then converted to flowrate over each day. These results along with the TDS values predicted by concentration correlation are shown by in Table **2**.

The objective of this case study is to design a mobile TMD system that can treat the flowback water of one well for the cycle of 14 days then moved to another well to treat its flowback water for the next 14 days and so on. The following constraints and data areto be considered:

A minimum recovery ratio of 75% is required, *i.e.*

$$\frac{\text{Treated Flowback Water (Cumulative Permeate)}}{\text{Cumulative Collected Flowback Water}} \geq 0.75 \tag{13}$$

The flared gases on site are utilized to pre-heat the TMD feed to 363 K

Table 2: Flowback Water Data Based on Regression Models

Day	Flowrate of Flowback W_{FB} (kg/s)	Mass Fraction of TDS, y_{FB}
1	16.68	0.028
2	7.89	0.035
3	4.62	0.042
4	3.28	0.050
5	2.54	0.057
6	2.08	0.064
7	1.76	0.072
8	1.52	0.079
9	1.34	0.087
10	1.20	0.094
11	1.09	0.101
12	0.99	0.109
13	0.91	0.116
14	0.84	0.123

The weight fraction of TDS in the reject should not exceed 0.35 (to avoid precipitation). A TDS mass balance around the TMD is given by:

$$W_{FB} y_{FB} = W_{\text{Reject}} y_{\text{Reject}} + W_{Permeate} y_{Permeate} \tag{14}$$

Assuming complete TDS rejection in the TMD (*i.e.*, $y_{Permeate}$=0) and using the definition of water recovery, ζ, as the ratio of the permeate flowrate to the feed flowrate, we get:

$$W_{\text{Reject}} = (1-\zeta) W_{FB} \tag{15}$$

we get:

$$\zeta = 1 - \frac{y_{FB}}{y_{\text{Reject}}} \tag{16}$$

Hence,

$$\zeta^{\max} = 1 - \frac{y_{FB}}{y^{\max}_{\text{Reject}}} \quad (17)$$

This constraint limits the extent of recovery for the high-TDS feeds (especially towards the end of the 14-day cycle). For the case study data and considering a maximum mass fraction of TDS in the reject as 0.35, the constraint given by Eq. (17) can be used to generate the data shown by Table **3**.

Table 3: Calculated Maximum Allowable Water Recoveries for the Case Study

Day	Mass Fraction of TDS, y_{FB}	Maximum Allowable Recovery
1	0.028	0.92
2	0.035	0.90
3	0.042	0.88
4	0.050	0.86
5	0.057	0.84
6	0.064	0.82
7	0.072	0.80
8	0.079	0.77
9	0.087	0.75
10	0.094	0.73
11	0.101	0.71
12	0.109	0.69
13	0.116	0.67
14	0.123	0.65

The TMD model developed by Elsayed *et al*. [6] is used in this case study. One of the key equations in the model is the permeate flux (J_w) equation:

$$J_w = B_w (p^o_{w,f} \gamma_{w,f} x_{w,f} - p^o_{w,p}) \quad (18)$$

where J_w is the permeate flux and B_w is the membrane permeability which is a function in the membrane properities as well as the membrane temperature. The terms $p^o_{w,f}$ *and* $p^o_{w,p}$ are the water vapor pressure of the feed and permeate. The term $\gamma_{w,f}$ is the activity coefficient of the water in the feed which may be calculated from concentration of the feed. For instance, in the case of NaCl the following expression [11] may be used to estimate the activity coefficient:

$$\gamma_{w,f} = 1 - 0.5 x_{NaCl} - 10 x^2_{NaCl} \quad (19)$$

where x_{NaCl} is the mole fraction of NaCl.

A direct contact polypropylene hollow-fiber TMD membrane MD020CP2N (manufactured by Microdyn) is used in this case study. The characteristics and properties of this membrane are reported by Al-Obaidani *et al*. [12] and Elsayed *et al*. [6]. For instance, the permeability is calculated through:

$$B_W = B_{WB} * T_m^{1.334} \quad (20)$$

where

1. $B_{WB} = 7.5*10^{-11} \frac{kg}{m^2.s.Pa.K^{1.334}}$ (21)

2. T_m is the average membrane temperature in K.

The following data for cost estimation are taken from Elsayed *et al*. (2013):

The annualized fixed cost of the TMD network, AFC_{TMD}, is given by:

$$AFC_{TMD} = 58.5 A_m + 1{,}115\, W_{FB} \quad (22)$$

where A_m is the area of the membrane in m^2 and W_{FB} is the flowrate of the flowback water fed to the TMD in kg/s.

The annual operating cost of the TMD network excluding heating (AOC_{TMD} in \$/yr) is given by:

$$AOC_{TMD} = (1{,}411 + 43 \times (1-\xi) + 1{,}613 \times (1+\upsilon)) \times W_{FB} \quad (23)$$

where ξ is the water recovery (ratio of permeate to feed for the TMD) and υ is the recycle ratio of the reject to feed.

By solving the multi-period optimization formulation at different recovery levels, the minimum cost solutions were obtained. A Pareto chart for the cost of permeate

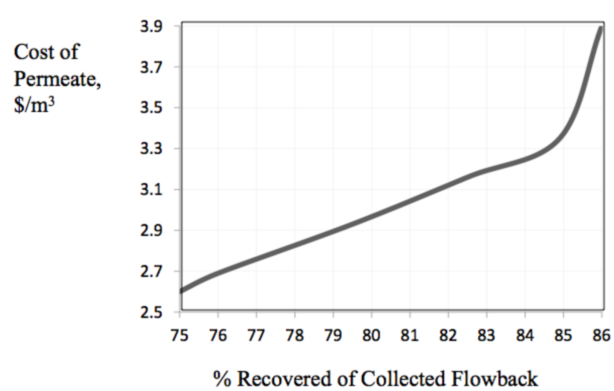

Figure 4: The Pareto Curve of Cost versus Recovery for the Case Study.

versus the percentage recovery of the flowback water as permeate is shown by Figure **4**. As can be seen, recoveries exceeding the minimum requirement of 75% entail an increase in the cost of permeate per m^3. This is attributed to the "capital productivity" factor. Initially, the whole area of the membrane system is used to treat the larger flowrates. As the flowrate of the flowback decreases over time, the needed membrane area becomes smaller. Therefore, the TMD system is not fully utilized later in the cycle but the fixed cost of the membrane is charged for the full size regardless of what fraction of the membrane area is used. The Pareto curve shows the tradeoff between cost and recovery. Depending on the relative importance of cost versus extent of water recovery, the decision maker should choose a point on the curve which reconciles both objectives.

CONCLUSIONS

A multi-period optimization formulation has been developed for the design of TMD systems that treat flowback wastewater in shale gas production. The devised approach is capable of handling the time-based fluctuations in flowrate and composition of the flowback streams. The TMD system is designed to be used for multiple wells. It enjoys flexibility and mobility while taking advantage of inexpensive low-level heating that is typically available in shale gas production sites. The optimization formulation accounts for operating and fixed costs of the system. It can also be used to trade off cost versus permeate recovery. A case study was solved for wells in the Marcellus Shale Play. The results of the case study indicate that flowback water can be treated in a cost-effective way using TMD and that conflicting objectives (such as cost and recovery) can be methodically handled using the devised approach. For 75% water recovery, the cost of the permeate is about \$2.6/m^3. As higher recoveries are sought, the cost per m^3 of permeate increases due to capital productivity factors in dealing with the decreasing amount of flowback water over time.

NOMENCLATURE

AFC_{TMD} Annual fixed cost of the TMD network, \$/yr

AOC_{TMD_NH} Annual operating cost of the TMD excluding heating, \$/yr

A_m Membrane area, m^2

B_w Membrane permeability, kg/m^2.Pa

B_{wB} Temperature-independent base value for the permeability, $\frac{kg}{m^2.s.Pa.K^{1.334}}$

$\overline{D}$ Design vector satisfying design constraints

D_t, Vector for design variables

H_t Number of operating hours in period t

Op_Cost_t Operating cost per hour

O_t, Vector for operating variables

$Permeate_{Desired}$ Total desired amount of permeate to be collected over the decision making time horizon

S_t Vector for state variables

T_m Average membrane temperature, K

$W_{FB,t}$ Flowrate of returned flowback at t period

$W_{Permeate,t}$ flowrate of the collected permeate during period t, kg/hr

W_{Reject} Flowrate of the reject, kg/hr

$x_{w,f}$ Mole fraction of the water in the feed

$y_{Desired}$ Desired composition of collected permeate

$y_{FB,t}$ Composition of returned flowback at t period

$y_{Permeate}$ Composition of Permeate

$y_{\text{Reject,t}}$ Composition of reject at cycle t

$y_{\text{limit,t}}$ Precipitation limit

Greek

$\gamma_{w,f}$ Activity coefficient of water in the feed to the TMD

τ_c Cycle time over which the flowback is treated

υ Ratio of recycled reject to raw feed, kg reject/kg raw feed

ζ Water recovery ratio, kg permeate/kg raw feed

Subscripts

f Feed stream to TMDN

u Hot stream

v Cold stream

w Water

t The set of periods within the decision-making time horizon

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