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Effect of pretreatment and operating conditions on the performance of membrane distillation for the treatment of shale gas wastewater

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ABSTRACT

The exploitation of shale gas has increasingly become important as a sustainable energy source but also has environmental issues including the production of wastewater with high total dissolved solids (TDS). Membrane distillation (MD) is a promising treatment option for such wastewater but may suffer from serious membrane fouling. Accordingly, this study examined the effect of pretreatment and operating conditions on the fouling behaviors of MD membranes. Hollow fiber membranes made of polyvinylidene fluoride (PVDF), polyethylene (PE), and polypropylene (PP) were compared in a laboratory-scale direct contact MD (DCMD) system. Real shale gas wastewater from oil and gas operations in Texas, United States was used. Results showed that the flux reduction ratio ranges from 13.6% to 27.7% to achieve 50% recovery ratio without pretreatment. Application of the pretreatments including FMX, flocculation – sedimentation (FS), and flocculation – sedimentation – microfiltration (FSMF) was found to be effective to retard MD flux decline. The characteristics of the fouling layers on the membrane surfaces were examined using scanning electron microscopy (SEM) and excitation emission matrix (EEM) analysis to understand the fouling mechanisms.

1. Introduction

Over the past few decades, unconventional oil and gas recovery has increased significantly in the United States and in many other parts of the world [1]. However, the increased intensity of activity and the proximity to populated areas has raised increasing public concern regarding the potential environmental effects and associated costs of hydraulic fracturing [1,2]. If unconventional production is conducted irresponsibly, groundwater and surface water may become rapidly polluted [3], and the supply of clean water is a very important future resource. Hydraulic fracturing involves the use of large quantities of water, proppants, and chemical additives that are injected into a shale formation under high pressure [3–5]. The resulting cracks in the formation release natural gas or oil, and horizontal drilling is used to increase the contact area between the structure and the fracturing fluid [5,6]. The major associated environmental issues include the outflow of chemicals from the fracturing fluid, surface runoff of chemicals and waste fluids, and failures of pressure control.

The wastewater generated by hydraulic fracturing contains a high concentration of salt, with total dissolved solids (TDS) in the range from 10,000 to 300,000 mg/L. Non-traditional natural gas extraction generates about 10 times more wastewater compared with conventional oil and gas extraction, e.g., the Marcellus Formation produces approximately 1.4 billion gallons of wastewater annually [7,8]. This wastewater generally has a high TDS content, with naturally occurring toxic compounds and a range of organic and inorganic fractions, including naturally occurring radioactive materials dissolved in the soil layer [9,10]. If this wastewater is discharged with inadequate treatment or without treatment, the high salinity and dissolved chemicals can pose a threat to the environment and to public health [11]. Accordingly, the disposal of these large volumes of wastewater is one of the most significant barriers to extended use of hydraulic extraction.

Existing treatment procedures include the capture and re-injection of the wastewater deep into the basement rocks [9,11]. Unfortunately, this process also raises environmental and geological problems, requires compliance with local regulatory systems, and incurs high trucking

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Table 1
Water quality parameters of shale gas raw wastewater and the pretreated water.

	Raw wastewater	FMX-B	FS	FSMF
Turbidity (NTU)	135	0.95	0.98	0.55
TDS (g/L)	120	93.3	90.0	87.0
SS (g/L)	0.16	–	–	–
pH	6.84	7.33	6.87	6.96
DOC (mg/L)	248.7	137.3	143.5	110.9
Color (Pt/Co scale)	2900	535	610	463
Na (mg/L)	49,057	43,361.2	46,372.2	42,611.5
K (mg/L)	302.4	270.1	283.7	260.8
Mg (mg/L)	383.6	254.6	262	250.4
Ca (mg/L)	3015.7	2552.4	2716	2503.7
F (mg/L)	13.5	–	1.07	–

costs [12–14]. Many alternative approaches have been proposed to this difficult-to-treat wastewater [15,16], including evaporation, nanofiltration (NF), reverse osmosis (RO), and osmosis used for seawater desalination [9,11,17]. However, NF and RO membranes are sensitive to scaling, particulate/colloidal fouling, organic contamination, extreme pH, and presence of oils, fats, insoluble liquids, and microbial biofilms [18,19]. In addition, the high pressure required to treat high concentration TDS such as shale gas wastewater makes treatment uneconomic. Conventional distillation such as multi-stage flash and multi-effect distillation is one alternative approach; however, the high energy usage has made it uneconomic. Intensive research is therefore being conducted on novel desalination technologies capable of producing clean, low-cost water [20–22].

One of desalination techniques that holds promise is membrane

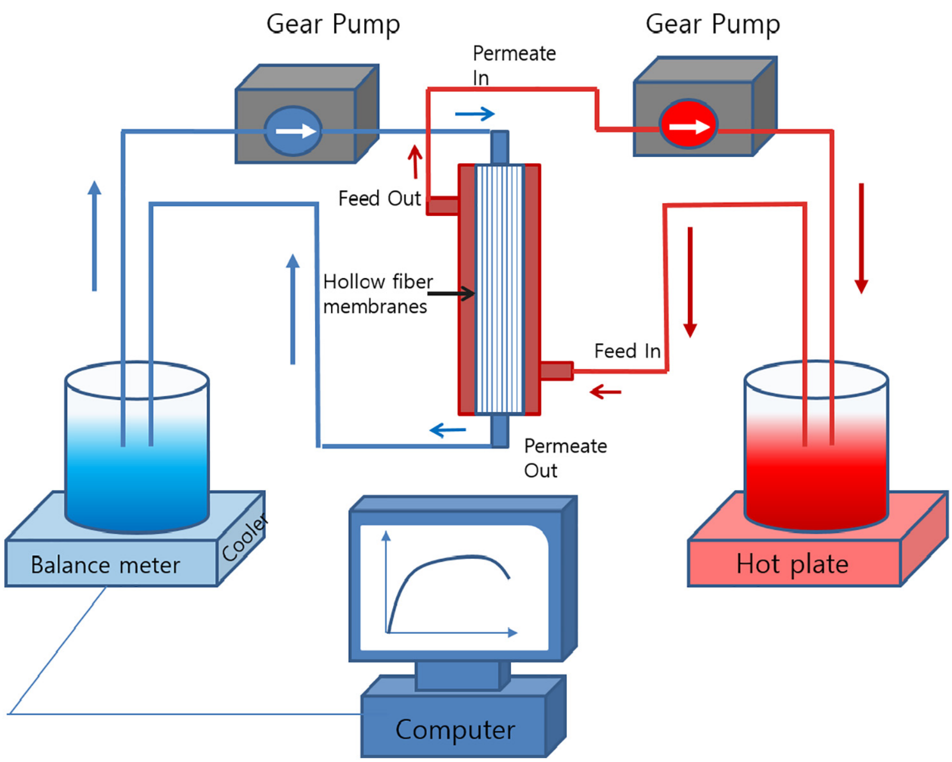

Fig. 1. Schematic of the laboratory-scale hollow fiber DCMD system.

Table 2
Experimental conditions for MD.

Conditions		Feed	Permeate
Operation type		Direct contact membrane distillation (DCMD)	
Flow rate		400 L/min	260 L/min
Temperature		50, 60, and 70 °C	20 °C
Membranes	PVDF membrane	Nominal pore size: 0.22 μm	Membrane area: 0.015 m^2
		Inner diameter: 0.7 mm	Flow direction: Outside-in
		Outer diameter: 1.3 mm	
	PE membrane	Nominal pore size: 0.4 μm	
		Inner diameter: 0.41 mm	
		Outer diameter: 0.65 mm	
	PP membrane	Nominal pore size: 0.22 μm	
		Inner diameter: 1.8 mm	
		Outer diameter: 2.6 mm	
Solution	Feed	Shale gas wastewater	(Eagle Ford, U.S.A.)
	Permeate	D.I. water	