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Desalination of hypersaline brines via Joule-heating: Experimental investigations and comparison of results to existing models

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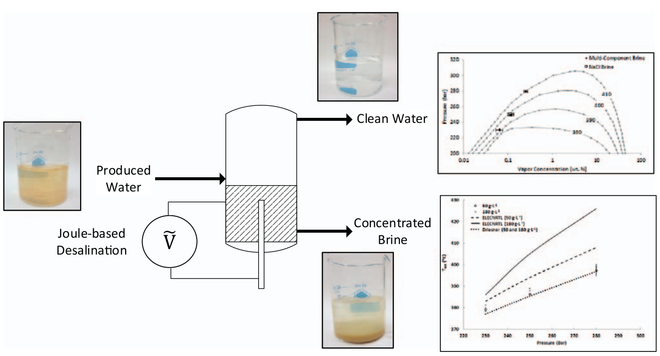
GRAPHICAL ABSTRACT



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ABSTRACT

Thermodynamic data of multicomponent brines are needed to properly treat brines from oil/gas and CO_2 sequestration operations. Joule-heating treatment of hypersaline brine is a unique methodology that allows for direct heating of brines without the need of external heating, potentially simplifying operation and reducing process footprint. The thermodynamic properties of multicomponent hypersaline brine (> 3.5 wt%) are unknown and limited to estimations based on single component brine data. Experimental data for multicomponent brines at elevated temperatures and pressures do not exist due to operating conditions above the pseudocritical point of pure water. This study combines experimental results for multicomponent brines using a Joule- heated desalinator at pressures of 230 to 280 bar and temperatures of 387 to 406 °C with thermodynamic models previously created for single component NaCl brines to identify the deviations resulting from the additional species. In addition, a comparison between an Aspen Plus® v9 simulation using the ELECTNRTL model with experimental results is provided.

1. Introduction

The ability to find adequate freshwater resources for energy production is a growing concern in the United States and around the world. A 2015 study completed by Konikow [1] showed that nearly all US aquifers have shown significant depletion since 1900, with a distinct increase in depletion rate during the 1940s and after the year 2000 [1]. While water resources are scarce in some locations, they are being contaminated in others.

Hydraulic fracturing is a water intensive process that also generates

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significant amount of produced water, also known as brine. A study conducted by Mielke [2] et al. showed that a single hydraulically fractured lateral can consume up to 5.6 million gallons of fresh water during the initial drilling and hydraulic fracturing phase of the well development. Another study conducted by Argonne National Laboratory estimated that the total volume of produced water in 2007 was nearly 882 billion gallons [3]. Further, as carbon emissions from power production have come under increased scrutiny interest in CO_2 sequestration via deep saline aquifers has increased. Projections indicate injection of one metric ton of CO_2 can produce 1 m^3 of saline water with total dissolved solids (TDS) concentrations ranging from 6 to 210 $g{\cdot}L^{-1}$ [4,5].

There have been many methods developed for decontaminating or disposing of hypersaline brines. These methods range from injection into underground reservoirs to treatment at municipal water plants [6]. Membrane technology including forward and reverse osmosis filtration is being pursued, but membrane fouling and pH sensitivity are still issues along with limited water recovery [7,8]. Simple distillation of the brines could be completed, but is highly energy intensive for large scale desalination of high salinity waste streams [8]. Evaporation pits are a simple technology that have been successfully used in desalination of low concentration brines, mainly from membrane desalination permeate, but require a large footprint and do not recover the desalinated water [8]. Mechanical vapor compression and multi-effect desalination systems are currently in use for desalination of produced water, but have low recovery ratios for high concentration brine feed water [9].

Ohio University (OHIO) is developing a supercritical-water based treatment process for hypersaline brines generated from industrial activities including oil/gas wells and CO_2 injection wells. Like many fluids, as pure water approaches its critical point, its enthalpy of vaporization (Δh_{vap}) diminishes dramatically, until the critical point is reached where a single phase is formed (i.e. no change of state occurs with heat addition). Utilizing supercritical water properties is a known means to desalinate brine waste generated by industrial processes [10–14]. However, most supercritical water reactors utilize externally heated designs, which possess high thermal lag, internal scaling issues, and high manufacturing costs. Due to these issues, such systems are unable to be cost-effectively scaled. To address this issue, new reactor designs which utilize alternative heating designs are being considered [15,16]. In this paper, Joule-heating, a result of the power dissipated as current travels through a non-ideal conductor, is evaluated for directly heating the brine. This novel design allows for direct control of power applied to the brine using the electrical conductivity of the brine when applying an alternating current (AC), resulting in a lower TDS content vapor product and a concentrated TDS liquid product. These streams will be referred to as vapor and liquid phases throughout the remainder of the document.

Early research into the properties of brines was driven by the need for thermodynamic data above standard temperature and pressure for use in geological modeling including groundwater [17,18] and sea floor hydrology [19,20]. The properties of high concentration brines under elevated pressure have been studied as early as 1931 [21], while elevated temperature and pressure test began as early as 1942 [17]. To date, numerous experimental studies have been completed for a variety of brines ranging in temperatures and pressures up to 646.2 °C and 4137 bar [21,17,22–25,10]. Several reviews of these and other experimental results have been conducted to develop models for the thermodynamic properties of brines at elevated temperature and pressure [18–20,24,26–29]. The results from these studies show that brine dissolved solids content has a distinguishable effect on brine density, enthalpy and specific heat.

Minimal experimental data was found regarding Joule-heating of or the Δh_{vap} for multicomponent hypersaline brines. The primary objectives of this study were to evaluate the ability to utilize direct electrical heating of hypersaline brines and develop information regarding thermodynamic properties associated with hypersaline brines with multicomponent dissolved solids (salt) compositions. Vapor-liquid equilibrium temperature (T_{VLE}) and enthalpy of vaporization were determined for two representative brine compositions at three pressures. Comparison of experimental results with a previously reported Aspen Plus® model [16] is also provided. Such results could allow for better process energy requirements for supercritical water treatment.

Table 1
Brine composition used in testing. Sodium added as sodium carbonate and sodium sulfate, followed by sodium chloride to complete the sodium balance. Chloride added as calcium chloride, barium chloride and strontium chloride, follow by sodium chloride to complete the chloride balance.

Ions	Brines		Ion source(s)
	50 $g{\cdot}L^{-1}$	180 $g{\cdot}L^{-1}$	
Na^+ ($mg{\cdot}L^{-1}$)	14,956	53,429	NaCl (> 99.0%), $NaHCO_3$ (> 99.7%), Na_2SO_4 (> 99.0%)
Ca^{2+} ($mg{\cdot}L^{-1}$)	4261	15,222	$CaCl_2$ (> 95%)
Ba^{2+} ($mg{\cdot}L^{-1}$)	27	97	$BaCl_2$ (> 95%)
Sr^{2+} ($mg{\cdot}L^{-1}$)	109	389	$SrCl_2$ (> 99%)
K^+	54	194	KCl (> 99.0%)
Cl^- ($mg{\cdot}L^{-1}$)	30,671	109,572	NaCl (> 99.0%), $CaCl_2$ (> 95%), $BaCl_2$ (> 95%), $SrCl_2$, KCl (> 99.0%)
HCO_3^- ($mg{\cdot}L^{-1}$)	82	292	$NaHCO_3$ (> 99.7%)
SO_4^{2-} ($mg{\cdot}L^{-1}$)	109	389	Na_2SO_4 (> 99.0%)
NH_4^+ ($mg{\cdot}L^{-1}$)	109	389	Ammonium hydroxide (28 wt%)
SiO_2 ($mg{\cdot}L^{-1}$)	10	34	Colloidal silica (40 wt%)
TDS ($mg{\cdot}L^{-1}$)	50,387	180,008	
Density ($kg{\cdot}m^{-3}$)	1032	1115	

2. Experimental and methodologies

2.1. Materials

All source materials were purchased from Fisher Scientific, with the exception of colloidal silica and ammonium hydroxide, which were purchased from Sigma-Aldrich and barium chloride, purchased from Reagents. The concentration and source materials for all constituents in the test brines are presented in Table 1. The test brine concentrations were selected based on a review of water produced by oil/gas and CO_2 injection wells [30–34]. All concentrations are presented in $mg{\cdot}L^{-1}$ or $g{\cdot}L^{-1}$ measured at normal temperature and pressure (293.15 K and 101.3 kPa). The multicomponent aqueous brine test brine was prepared at a concentration of 180 $g{\cdot}L^{-1}$ in a 25 gal tank circulating for a minimum of 12 h using a 3250 GPH Hydor Koralia Magnum pump. After mixing was complete, precipitates were filtered using a 0.35 μm pleated cartridge filter. The 50 $g{\cdot}L^{-1}$ brine was prepared by diluting the premixed 180 $g{\cdot}L^{-1}$ brine by a factor of 3.6 using DI water. Density of the 50 and 180 $g{\cdot}L^{-1}$ test brines were calculated using NaCl data from EES [35], and found to be 1032 and 1115 $kg{\cdot}m^{-3}$ with salt content of 4.9 wt% and 16.1 wt%, respectively.

2.2. Desalination system

A supercritical water test system using Joule-heating for treatment of brines was developed with the aim of investigating the proposed water management process. A process and instrumentation diagram of the prototype test system and a cross section view of the desalinator are shown in Fig. 1. The system utilizes a high pressure liquid chromatography pump (P-100) to supply a flow rate up to 300 mL per minute. The brine was pumped through a high pressure tube in tube heat exchanger (HX-100) to recover heat from the processed fluid, followed by a preheater (HX-101) to control the desalinator inlet temperature. The desalinator has a radial electrode configuration with an inner electrode diameter of 0.25 in., and is constructed from Hastelloy C-276. The 1 in.