



Electrodialysis: An application for the environmental protection in shale-gas extraction



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ABSTRACT

Water is one of the resources used to fracture the host formation and retrieve the trapped gas during the process of shale-gas extraction. A portion of the injected water, which is known as flow-back water, is recovered during the initial extraction of the gas. The main characteristic of flow-back water is the very high concentrations of dissolved solids (e.g. calcium, sodium, and chloride), which renders its treatment highly challenging for most existing treatment methods. An alternative for onsite treatment (or pre-treatment) and recycling of the flow-back water is electrodialysis. The method is based on an electrically assisted membrane process for separation of salts from the solution. In this study, the efficiency of the electrodialysis for desalination of flow-back water was investigated at the laboratory scale using samples of flow-back water from the Marcellus shale-gas formation.

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1. Introduction

Shale formations are known to have extremely low permeability, which makes hydraulic fracturing (fracking) a required process for economically viable shale gas extraction [1]. The process consists of the injection of the fracking fluid at high pressure into the formation. The fracking fluid is a combination of water and various chemical additives [2]. The volume of the injected fracking fluid varies among different shale gas formations and even among the wells of the same shale gas formation. In the Marcellus formation, the volume of injected fracking fluid ranges between 10,000 and 30,000 cubic meters per well [3,4]. Almost 90% of the injected volume remains bound to the dry shale matrix and only 10–20% is recovered as a wastewater stream, known as flow back water [2–5]. The main supplies of water for hydraulic fracturing are surface water, ground water, and recycled water [2].

Flow-back water is considered as a mixture of fracking fluids returning to the surface and chemical constituents originating from the shale formation [2,4]. The predominant constituents of flow-back water are the dissolved salts [6]. The dissolved salts can be identified using conductivity, salinity, or total dissolved solids (TDS) measurements. The concentration of Total Dissolved Solids (TDS) in the flow back water of the Marcellus formation varies between 30,000 mg/L and 200,000 mg/L [1,2,7,8]. The most

common compounds of TDS are calcium, magnesium, chloride, sodium, sulfate, barium and iron [1,2,6,7]. The flow back also contains chemical additives used during the drilling, fracturing, and operation of the well and in some cases naturally occurring radioactive materials [1,2,7]. With a production of about 21 billion barrels per year (2011) in the USA, flow-back water is the largest byproduct stream of shale-gas extraction [2]. The high salinity requires proper management of the flow back water [6]. In Marcellus, the gas industry drilled 1386 Marcellus wells in 2010, compared to 763 in 2009. From 2012 to date, 2013 permits were issued and 1074 wells were drilled. Therefore, the water demand, and consequently, the volume of the wastewater stream has increased significantly in recent years [3,4]. Although, the development operations in the Marcellus have access to sufficient water supplies, the treatment and disposal of the flow-back water represent a major cost item [1,5,9].

The common practice in shale-gas extraction for the management of flow-back water is the disposal in deep wells (Class II wells). In the Marcellus, there are only 8 Class II wells for this purpose [1]. In 2013 in Pennsylvania, about 87% of the flow-back water was transported to commercial wastewater treatment plants that operate under the supervision of the state environmental protection agency [1,2,5,8]. However, after the recent earthquakes in the state of Ohio that are linked with the injection of waste water in deep wells and the recent environmental indicators of elevated TDS have been noted in the Monongahela River (Pennsylvania and West Virginia), including elevated concentrations of chloride and golden algal blooms that produce toxins that can suffocate aquatic

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organisms, the need for treatment and recycling of the flow back water has become more apparent [6,8]. A step forward has been made on this direction as the amount of flow back water that is recycled has increased from 11% prior 2011 to 56% in 2011 [8].

Incidents like the above-mentioned render the treatment of the flow back water vital for the protection of the public and of the environment. The industry standard for acceptable recycle water TDS level, is a maximum of 50,000 mg/L [10]. However, limits for acceptable reuse may vary widely depending on the driller. In some cases water with a TDS level greater than 50,000 mg/l has been used as recycle water [10]. Most companies apply a basic settling and filtration treatment of the flow back water, which is subsequently mixed with fresh water in order to reduce the TDS levels so that it can be reused for other fracturing jobs. However, this method requires the use of additional fresh water [2,5]. Since May 2010, the state of Pennsylvania has established more stringent discharge requirements for the TDS concentrations and the shale-gas industry is in search of alternative efficient and economical remediation technologies [5].

Various technologies have been developed and tested over the past years for treatment of flow-back water. Examples of those technologies are thermal treatment, membrane treatment, and various hybrid and advanced treatment technologies. Most of these technologies are unable to treat wastewater with high TDS levels [1,2,11]. Therefore, a need for the development of pre-treatment methods to reduce the TDS level has arisen. A membrane treatment technology called electrodialysis could be a potentially promising alternative for the reduction of TDS levels [1,9]. This paper investigates the efficiency of electrodialysis as a desalination process (pretreatment) at the laboratory scale for flow-back water samples taken from Marcellus shale wells. The goal of this pre-treatment method is the reduction of the TDS level to an amount that is manageable by the currently used treatment methods. The main focus of the study is the investigation of the parameter which affects the most the process in terms of TDS removal.

2. Background

Electrodialysis was first proposed in 1890 by Maigrot and Sabates for demineralization of sugar syrup [12]. Electrodialysis has been used for over 50 years for desalination of brackish water, seawater and industrial water [13–21]. It is also used in the food industry for purification, modification, or concentration of food [22], and recently for the production of organic acids [23]. Electrodialysis has been tested for the desalination of coal-mine brine and produced water with encouraging results on its energy consumption when compared with other methods [24,25]. The method is an electrochemical process in which the ions are transferred through ion-selective membranes from one solution to another using a low Direct Current (DC) electric field as the driving force [14,26]. Two different outflows are produced in the process, the dilute flow with lower TDS level than the inflow, and the concentrated flow with higher TDS level than the inflow [1,14,27–29]. Fig. 1 is a schematic showing the operating principle of electrodialysis. The performance of the electrodialysis method depends on various parameters such as the applied voltage, the flow rate, the properties of the membranes, the composition of the inflow, and the design parameters of the equipment (e.g. stack construction, cell dimensions, etc.) [17,18,26,27,30,31]. The main investment cost of the method is that associated with the membranes, which depends on the required membrane area. The cost of operation is mainly determined by the energy requirements for the application of the DC electric field [30].

Ion selective membranes are made of a macromolecular material having ionizable groups such as ion-exchange resins [22].

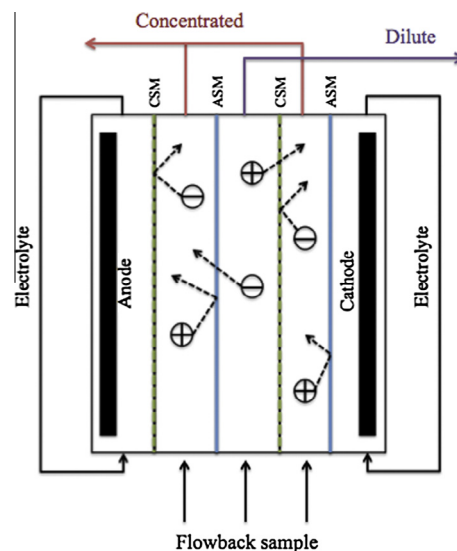


Fig. 1. Schematic of the principle of Electrodialysis (CSM: Cation Selective Membranes; ASM: Anion Selective Membranes).

The ion selective membranes are basically ion exchange resins in the form of a film. They have evolved from a laboratory tool to an industrial product [29,32,33]. The two main categories are the cation and the anion selective membranes. The cation selective membranes include negatively charged groups (e.g. $-\text{SO}_3^-$, $-\text{COO}^-$, $-\text{PO}_3^{2-}$) that only allow the movement of the cations. In contrast, the anion selective membranes allow the movement of the anions using positively charged groups (e.g. $-\text{NH}_3^+$, $-\text{NRH}_2^+$, PR_3^+). The desired properties of the ion selective membranes are high permselectivity; low electrical resistance; high mechanical, thermal, and chemical stability; and low cost of production [29,32,33].

3. Experimental program

3.1. Experimental set-up

The experimental set up used in this study is shown in Fig. 2(a). It consists of the electrodialysis device, the peristaltic pump, the power supply for application of the DC electric field, and the container for the electrolyte. The design of the device is based on the principle of electrodialysis as shown in Fig. 1. Three pairs of anion and cation selective membranes are used within the device. The role of the central pair of membranes is to trap the anions and the cations of the feed (flow back water) in the concentrated chambers. The two external pairs of membranes are used to separate the electrolyte from the feed.

The device consists of five chambers including two electrode chambers, two chambers for the concentrated outflow, and one chamber for the dilute outflow as shown in Fig. 2(b). The chambers are made of propylene to prevent reactions caused by application of the electrical field. The dimensions of the device are 30 cm × 9 cm × 9 cm. For the tests that used sodium sulfate (Na_2SO_4) as the electrolyte, titanium electrodes are placed in the chambers using four titanium bolts that are also used for the connection of the electrodes to the power supply. However, electrodes made of graphite are used for the tests using sodium chloride (NaCl) as the electrolyte, due to the corrosion that the NaCl cause to the titanium. Bolts that penetrated the frame of all the chambers are placed in the perimeter to keep the device sealed and to prevent leakage. A peristaltic pump is used for injection of the feed to the three central chambers and then for circulation of the electrolyte within the electrodes chambers. Flow only exists

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