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Treatment and reuse of shale gas wastewater: Electrocoagulation system for enhanced removal of organic contamination and scale causing divalent cations

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ABSTRACT

The present study explores the feasibility of using the electrocoagulation (EC) process for the treatment and reuse of wastewater produced during shale gas recovery by hydraulic fracturing. The electrocoagulation process has been evaluated for the removal of suspended solids, total organic carbon (TOC) and scale (hardness) causing divalent cations, which, if untreated, can clog the gas well. Experiments were performed with actual shale gas wastewater (ASWW), synthetic shale gas wastewater prepared with low concentration of dissolved salts (SSWW – LDS) and synthetic shale gas wastewater prepared with a high concentration of dissolved salts (SSWW – HDS).

EC is found to be effective for removing TOC and hardness from both the actual and synthetic shale gas wastewaters. The electric energy required per unit mass (E_{EM}) for removal of TOC for ASWW, SSWW – LDS and SSWW – HDS are 243, 102 and 70 kWh/kg respectively. The E_{EM} for removal of hardness for ASWW, SSWW – LDS and SSWW – HDS are 303, 104 and 25 kWh/kg respectively. The high conductivity of SSWW – HDS helps in achieving higher currents and hence the lower reported E_{EM} values for SSWW – HDS. Also, under alkaline conditions, the performance of EC increases significantly. Combination of aeration with EC is also found to increase the performance of EC, especially for wastewater containing high concentrations of chloride ions.

1. Introduction

The increase in water demand due to population and industrial growth is stressing the existing fresh water resources and causing physical and economical water scarcity. Recently, the use of large volumes of fresh water and generation of large volumes of highly contaminated wastewater during the production of shale gas has attracted a lot of attention from environmental groups as well as from the general public [1–7].

Shale gas is the natural gas trapped in impervious sedimentary rocks about 6000–10,000 feet deep from the earth's surface. Earlier, it was thought to be difficult to extract this trapped gas but technological advancements in horizontal drilling and hydraulic fracturing have made commercial extraction of shale gas economical. During hydraulic fracturing large volumes of water with proppant (sand) and chemicals are injected at high pressure down and across horizontally drilled wells as deep as 10,000 feet below the surface to create cracks and fissures in the rock formations and release the natural gas trapped in them. Part of the injected water gets trapped in the shale formation while the remaining portion of the injected water along with natural water trapped in the underground formation flows back to the surface once the pressure in the well is released.

The wastewater produced contains a high concentration of dissolved salts (5000–250,000 mg/L) which is about 6 times higher than the salt content of sea water. It also contains suspended solids (0–3000 mg/L), oil (5–1000 mg/L), divalent cations (calcium: 0–20,000 mg/L, barium: 0–10,000 mg/L, strontium: 0–5000 mg/L, magnesium: 0–2000 mg/L), monovalent cations (sodium: 2000–100,000 mg/L and potassium: 0–750 mg/L), anions (chloride: 3000–200,000 mg/L, sulfate: 0–5000 mg/L, carbonate: 0–1000 mg/L and bicarbonate: 100–6000 mg/L), BTEX (Benzene, Toluene, Ethylbenzene and Xylene) (0–100 mg/L), bacteria (0–10^5

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MPN/100 mL) and sometimes, radioactive elements (0–1000 pci/L) [5,8–10].

Considering the high concentration of contaminants, shale gas wastewater poses potential environmental threats and, if released into surface and underground freshwater sources, can affect the health of people, aquatic life and agriculture. The contaminants can also deplete oxygen in water and can react with disinfectants in water treatment plants to form cancer causing compounds [11,12]. The large volumes of produced wastewater is stored at the well site in open impoundment pits and tanks and then disposed by transportation or via a pipeline. The improper construction of impoundment pits allows the contaminants to leach into the soil into underground freshwater aquifers. For example, in Pavillion, Wyoming (US), high concentrations of benzene, xylene and other organic compounds were found in shallow wells near impoundment pits [13]. Considering its potential health and environmental impact, the shale gas industry in the US is prohibited from discharging produced water into freshwater streams and shale gas wastewater is also prohibited for being treated at municipal wastewater treatment plants. The US EPA is also developing new rules for proper disposal of produced water. Currently, produced water is disposed of into the deep injection wells which are regulated by the US EPA. The disposal cost ranges from 1 to 3 USD per bbl (6.3–18.9 USD per m^3) [14]. However, only a limited numbers of deep injection wells are located close to shale gas extraction sites. Major shale plays in the US such as the Marcellus shale play don't have suitable geological conditions for construction of deep injection wells. Hence, well operators have to transport wastewater over long distances before they can dispose it off into deep injection wells. The average wastewater transportation cost in the two major shale plays 'Barnett' and 'Marcellus' is in the range of 1–3 USD/bbl (6.3–18.9 USD per m^3) and 8–19 USD/bbl (50.3–119.5 USD per m^3) respectively [14].

Hence, in order to sustain growth, one of the main challenges facing the shale gas industry is of reducing the consumption of freshwater as well as minimizing the adverse effects of produced wastewater on the environment. The best way to solve this problem is to treat the produced wastewater at the shale gas production site and reuse it for applications such as drilling and hydraulic fracturing at the same well or at a nearby well [15–17]. This will help in minimizing the stress on the community's fresh water resources and help to minimize expenses on transportation and disposal of wastewater to deep injection wells. It will also reduce the number of truck trips for transportation of fresh water and wastewater and hence help in minimizing pollution.

In this study electrocoagulation (EC) is evaluated as a candidate technology for the treatment and reuse of shale gas wastewater, as it can remove fine suspended particles [18,19], microorganisms [20,21], heavy metals [22–24], oil and grease [25], organic matter [26,27] and hardness-causing ions [28]. It has high water recovery, produces less sludge and has lower environmental impact [29]. Treatment equipment based on electrocoagulation will be compact and more suitable for on-site use. However, electrocoagulation requires significant amounts of electricity. Considering the high concentration of contaminants present in shale gas wastewater, it will make the treatment process more expensive. Moreover, it is less-effective against the removal of dissolved organic contamination and hardness-causing divalent cations [30]. Hence, the objective of this work is to evaluate the feasibility of using electrocoagulation for the treatment of shale gas wastewater containing varying concentrations of different contaminants. Another objective of the current study is to increase the performance of electrocoagulation for the removal of organic contamination and hardness causing cations.

2. Materials and methods

2.1. Wastewater collection from shale gas well site and preparation of synthetic shale gas wastewater in the laboratory

2.1.1. Wastewater collected from shale gas well site

Wastewater was collected from the shale gas well site from the inlet pipe of the wastewater pond, on the 1st and 2nd days after hydraulic fracturing (the site details are not given here to maintain confidentiality of the company). Six samples of 5 L each were collected and mixed in equal volumes to create a representative sample which was used for further experimentation. The physicochemical parameters of the six samples and their mixture (sample 7) are shown in Fig. 1. As seen in Fig. 1, the wastewater samples collected after different time intervals show wide variation in their physicochemical parameters. The pH of the sample varied from 5.9 to 7.5, the conductivity varied from 4000 to 27,000 μS/cm, the total organic carbon (TOC) varied from 45 to 1251 mg/L and hardness varied from 140 to 280 mg/L as $CaCO_3$.

2.1.2. Synthetic shale gas wastewater

Synthetic shale gas wastewater was prepared by adding three major contaminant groups to de-ionized water: suspended solids, organic contaminants and dissolved salts. Wastewater from shale gas wells is found to span a wide range of total dissolved salts (TDS) values depending upon the shale play and the life time of the well. The TDS of shale gas wastewater from the Barnett shale gas play (US) is in the range of 23,600–98,600 mg/L, whereas in the Marcellus shale gas play (US), it is in the range of 8500–238,000 mg/L [31]. Esmaeilirad et al. observed an increase in TDS with time after hydraulic fracturing [8]. For example, a shale gas well in the Wattenberg field of Northeastern Colorado recorded a TDS value of 12,593 mg/L on the first day after hydraulic fracturing which increased to 38,174 mg/L on the 161st day after hydraulic fracturing [8]. In order to capture the wide variations in TDS of shale gas wastewater in future experiments, two types of synthetic wastewaters were prepared, one with a low concentration of dissolved salts (resulting in low hardness and low conductivity) (Conductivity ~14,000 μS/cm) and a second with a high concentration of dissolved salts (resulting in high hardness and high conductivity) (Conductivity ~113,000 μS/cm). The synthetic shale gas wastewater was prepared in three stages. In the first stage, dissolved salts were added. In the second stage, crude oil and surfactant were added to make a stable oil-in-water emulsion. In the third stage, fine test dust was added as a source of suspended solids.

2.1.2.1. First stage – addition of dissolved salts. Anhydrous calcium chloride $CaCl_2$ (Make-Thomas Baker) and sodium bicarbonate $NaHCO_3$ (Make-Thomas Baker) were used as the source of permanent hardness (non-carbonate) and temporary hardness (carbonate) respectively. Sodium chloride NaCl (Make-Thomas Baker) was used as the source of monovalent cation and chloride. The amounts of chemical compounds/substances added in the DI water for preparation of synthetic shale gas wastewater with low concentration of dissolved salts (SSWW – LDS) and synthetic shale gas wastewater with high concentration of dissolved salts (SSWW – HDS) are given in Table 1.

2.1.2.2. Second stage –addition of organic contamination (crude oil). In order to make a stable oil-in-water emulsion, a surfactant was added to the solution prepared in the first stage and mixed using a homogenizer (Make-Snowtech Process Equipment, Mumbai, India). Subsequently, crude oil (Density 822.9 kg/m^3 at 288 K and Viscosity 0.00302 Pa S at 313 K) was added to this solution while continuing mixing using the homogenizer. Since surfactants having HLB (hydrophilic-lipophilic balance) in the range of 8 to 18