



## New fouling prevention method using a plasma gliding arc for produced water treatment



K.C. Wright<sup>a</sup>, H.S. Kim<sup>a</sup>, D.J. Cho<sup>b</sup>, A. Rabinovich<sup>b</sup>, A. Fridman<sup>a,b</sup>, Y.I. Cho<sup>a,b,\*</sup>

<sup>a</sup> Department of Mechanical Engineering and Mechanics, Drexel University, Philadelphia, PA, USA

<sup>b</sup> Drexel Plasma Institute, Drexel University, Camden, NJ, USA

### HIGHLIGHTS

- New plasma-based method is introduced to prevent mineral fouling in produced water.
- Plasma arc discharge removed bicarbonate ions in produced water.
- CaCO<sub>3</sub> fouling was prevented in heat transfer surface with zero bicarbonate ions.
- Excessively high Ca<sup>2+</sup> ions in water did not cause CaCO<sub>3</sub> fouling problem.

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### ABSTRACT

The objectives of the present study were to test a new plasma-based method for bicarbonate removal in water produced from hydraulic fracturing for shale gas and to verify bicarbonate removal as a mechanism for mineral fouling prevention. The study employed dual hypotheses: (1) gliding arc plasma can reduce bicarbonate concentration of produced water to negligible levels, and (2) water without bicarbonate content is unable to cause mineral fouling in the form of calcium carbonate scale even in the presence of extremely high concentrations of calcium ions as is often found in produced water. The plasma treatment of produced water reduced the concentration of bicarbonate ions from a range of 684–778 ppm to zero. Subsequently, heat transfer fouling tests were conducted with water samples, where the concentration of bicarbonate ions varied from 0 to 500 ppm with a fixed concentration of calcium ions of 5000 ppm. The weight of mineral scale that accumulated on a heating element over time was determined. The present test results showed that no fouling was formed over 30 h on the heating element in the water without bicarbonate ions. This proof-of-concept study demonstrated that plasma treatment is capable of reducing the concentration of bicarbonate ions in produced water to a level where CaCO<sub>3</sub> fouling can be prevented on a heat transfer surface. The plasma gliding arc may represent a new non-chemical fouling prevention method for the treatment of produced water.

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### 1. Introduction

Natural gas production through hydraulic fracturing of shale formations is a rapidly accelerating field. Shale gas is now widely viewed in the U.S. as a key asset, which can bolster a more energy-independent future. In the midst of widespread optimism towards shale gas, certain environmental concerns have arisen that center on seismic effects of hydraulic fracturing as well as the aggressive consumption of water and, more importantly, possible pollution of local water resources by produced and flowback waters.

Produced water is conventionally treated using a range of physical, chemical, and biological methods. Since there are multiple needs that

should be addressed in produced water treatment, a variety of different conventional methodologies have been used, as recently reviewed by Ahmadun et al. [1], including the following: activated carbon, various forms of filtration (such as sand filters, cartridge filters, multi-media filtration, membrane filtration), organic-clay adsorbers, chemical oxidation, UV disinfection, chemical biocides, air strippers, chemical precipitation, water-softening by the application of lime soda, clarifiers, settling ponds, ion exchange, reverse osmosis, evaporation, steam stripping, and acidification. In nearly all of the above cases, each modality of technology typically achieves a single treatment target. For example, UV disinfection can only destroy bacteria and is unable to address any of the other objectives. Similarly, Hayes and Arthur [2] described a number of processes that are potentially applicable to produced water treatment, indicating that each process can be applied to only a limited number of basic functions.

Water hardness consists of permanent (i.e., calcium) hardness and temporary (bicarbonate) hardness [3]. In many industries, the removal

\* Corresponding author at: Department of Mechanical Engineering and Mechanics, Drexel University, Philadelphia, PA, USA. Tel.: +1 215 895 2425; fax: +1 215 895 1478. E-mail address: [choyi@drexel.edu](mailto:choyi@drexel.edu) (Y.I. Cho).

of temporary hardness by the addition of lime (i.e., CaO or Ca(OH)<sub>2</sub>) is a common practice [3–5], and lime-softening of water is a highly effective chemical means for preventing fouling. The drawback of adding lime is that it increases the solid content of CaCO<sub>3</sub> sludge, in effect increasing the overall cost of disposal to landfills or landfarms, which is an important economic and environmental driver in the treatment of produced water [1].

The plasma used in this study is ionized gas having concentrated energy with highly localized temperature increases near the arc of the gliding arc discharge (GAD) [6]. In particular, the present study focuses on the ability of the GAD to prevent mineral fouling in produced water by removing bicarbonate ions in produced water. Although oil field produced waters have been reported to contain a large amount of bicarbonate, with concentrations of 77–3990 mg/L, the amount of calcium ions, with maximum concentration of 25,000 ppm, is significantly larger than that of the bicarbonate ions [7]. Hence, it is prudent to remove the bicarbonate ions rather than the calcium ions in the produced water. Table 1 includes data on bicarbonate concentration for gas field produced water samples used in the present study, obtained from the Marcellus Shale in Pennsylvania.

The geometry of the GAD system and its ability to distribute plasma in water makes it well-suited to alter water chemistry, including the bicarbonate ion concentration, in water. Although the present study focuses on the ability of the GAD to prevent mineral fouling in produced water, it has been well reported in literature, the ability of the GAD to also oxidize hydrocarbons including partial oxidation of methane [8], and fuel conversion and reforming [9,10], as well as GAD ability to inactivate bacteria in water [11–13], especially for *Escherichia coli* in water.

Recently we reported that the GAD produced H<sup>+</sup> ions in water, thereby reducing the pH of water [14,15]. The H<sup>+</sup> ions react with bicarbonate ions in produced water, converting them to H<sub>2</sub>O and CO<sub>2</sub> (gas) [3]. Detailed geometry and specifications of the GAD used in the treatment of produced water have been reported elsewhere [14,15] and are not repeated here.

Since the removal of temporary hardness using plasma discharges has not been previously reported in scientific literature or in industry publications, it is necessary to demonstrate the efficacy of calcium carbonate scaling prevention in plasma-treated produced water. Therefore, the purpose of this study was to investigate the feasibility of a new method for preventing scaling in produced water through the removal of bicarbonate ions. The study was conducted in two parts, each with their own objectives, as shown in Fig. 1: (1) to test a plasma-assisted method for bicarbonate removal in produced water, and (2) to verify bicarbonate removal as a mechanism for scale prevention. For the first objective, a plasma gliding arc discharge (GAD) [6,16–18] was applied to produced water samples from hydraulic fracturing to demonstrate the softening of water and removal of bicarbonate content. To meet

the second objective, high concentrations of calcium ions and bicarbonate ions ranging from zero to 500 ppm were artificially added to distilled water or municipal water. By applying heat over 30 h to evaporate water, the effect of bicarbonate removal was verified as a mechanism for CaCO<sub>3</sub> fouling prevention. This pilot study was designed to demonstrate a new method using plasma GAD for pretreatment of complex waste waters for the purpose of decreasing bicarbonate concentration and mineral fouling mitigation even in the presence of high calcium ion concentrations.

## 2. Experimental methods

A plasma gliding arc discharge configured for reverse tornado flow was utilized to generate air plasma which was applied to produced water for treatment. The plasma GAD reactor consisted of a 1-L reservoir and a GAD plasma generator connected to a power supply and a compressed air line. Feed water entered the reactor using a peristaltic pump. For safety, a snorkel ventilation unit was employed directly above the setup for safe extraction of air and vapor from the system to outside of the laboratory. For the first part of this study, produced water samples from hydraulic fracturing for shale gas were used (Springville, PA). The compositions of the water samples used from this Marcellus shale rock formation are as follows: TDS of 36,000–38,000, calcium ion concentration of 940 mg/L–1010 mg/L, potassium concentration of 1870–2080 mg/L, sodium concentration of 10,600–11,300 mg/L, pH of 7.1–7.71, and bicarbonate concentration of 685–778 mg/L.

At the beginning of each test, 700 mL of produced water was isolated into a 1-L beaker. At baseline, 50 mL of this sample was extracted with a syringe, and laboratory assessments including alkalinity and pH measurements were performed. The remaining 650 mL volume was loaded into the cylindrical plasma reactor, and plasma power was turned on. The power supply parameters for generation of plasma were recorded, including voltage and current. After 10 min, plasma power was turned off, and 50-mL of treated water was sampled for laboratory assessments including bicarbonate ion concentration. These tests were repeated a total of six times and reported in Table 1.

For six additional tests, plasma treatment was continued beyond 10 min until the bicarbonate concentration was reduced to zero. Water samples of 50-mL volume were extracted every 10 min for 1 h. Laboratory assessments were performed on all extracted water samples to assess the time required to achieve bicarbonate removal (i.e., time to zero bicarbonate) as reported in Table 1. During testing water was recirculated at a flow rate of 50–200 mL/min using the peristaltic pump. Bicarbonate concentration was determined based on the assumption that alkalinity was due almost entirely to hydroxides, carbonates, or

**Table 1**  
Plasma-induced bicarbonate removal in various produced water samples.

Test no.	Volume (mL)	Plasma treatment time (min)		Time to zero bicarbonate (min)	Voltage (kV)	Current (A)
		0	10			
Bicarbonate concentration (ppm)						
1	650	699	467	–	2.1	0.1
2	650	685	475	–	2.1	0.1
3	650	778	560	–	2.1	0.1
4	650	745	370	–	2	0.1
5	650	685	458	–	2	0.1
6	650	685	203	–	1	0.2
7	650	760	364	50	2.1	0.1
8	650	747	347	30	2	0.1
9	650	684	317	30	2	0.1
10	650	752	472	30	2	0.1
11	650	685	292	30	2	0.1
12	650	685	83	20	2	0.1

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