



System-level model and experiments for irrigation water alkalinity reduction and enrichment using an atmospheric pressure dielectric barrier discharge

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ABSTRACT

The treatment of distilled water with varying amounts of dissolved sodium bicarbonate (representing alkalinity) is considered using an atmospheric pressure electrical discharge. The discharge ignited between a capillary tube (used as powered electrode) and a ground electrode wrapped around the beaker holding the treated water consists of streamers propagating in ambient air and striking the water surface. The streamer interaction with water is shown to lead to a decrease in pH and an increase in nitrate concentration. The pH variation with time is shown to be similar to a titration curve for acid-base neutralization with final pH values around 3 for 22 min of treatment. The nitrate ion concentration increase with time is consistent with a two-rate system-level model that is characterized by two asymptotic rates for NO_3^- creation by the plasma. The two asymptotic rates are calibrated to be about $2.7 \mu\text{mol/min}$ and $22.5 \mu\text{mol/min}$ with the transition between the two rates occurring at the breakeven time that is representative of the time required for all dissolved bicarbonate to be consumed by the plasma treatment. The increase in rate of NO_3^- creation at the breakeven time is attributed to the increase in conductivity of the treated solution once all bicarbonate is consumed thereby modifying the plasma properties. Another system-level model that is based on the observed pH variation is also considered for comparison with measured data. While both system-level models have some discrepancies with the measurements, the two-rate model based on the nitrate ion concentration is concluded to be more useful for determining the NO_3^- formation rates in the context of irrigation water enrichment. The discrepancies are attributed to the simplicity of the system-level models considered here where the effect of the plasma is completely represented by the creation of just one chemical species in HNO_3 thereby neglecting potentially important species such as HNO_2 and H_2O_2 . Nevertheless, the proposed system-level model could greatly assist in the design of plasma treatment systems with specified alkalinity, pH and nitrate ion levels for irrigation water.

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1. Introduction

The rapidly growing world population, with an estimated 11 billion in 2100 (Gerland et al. (2014)), has led to a great emphasis on food security and food production as two major problems that need to be tackled. This, in turn, has contributed to an increased importance of irrigation water management and the need to potentially grow crops in regions that do not have suitable soil or water resources. The acidity or alkalinity of the irrigation water (Valdez-Aguilar and Reed (2007)) and soil is an important parameter

that determines the suitability for agriculture. Specifically, pH measures the concentration of hydrogen ions in the water/soil and should be between 5.5 and 6.5 for most crops to ensure good irrigation water quality (Simsek and Gunduz (2007)). The soil and irrigation water pH play a crucial role in determining the outcomes of plant growth (Zebarth et al. (2015)). The alkalinity, determined by the concentration of bicarbonate and carbonate ions (and expressed in units of ppm or mg/L or milliequivalents/L), should also be limited to ensure healthy crops. While the tolerable levels of alkalinity are specific to the crop and the container size, a general rule of thumb is to limit the bicarbonate levels to 120 mg/L and carbonate levels to 15 mg/L respectively. While the source of irrigation water is region-specific, the United States Geological Survey (USGS) website

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indicates Dieter et al. (2018) that, out of the 118,000 million gallons per day of irrigation water 60,900 million gallons (52%) per day came from surface water with the remaining coming from ground water. The principle source of alkalinity in both surface and ground water is the carbon dioxide present in atmosphere. In particular, unsaturated soil zone (subsurface region between land and water table) air can contain significant amounts of carbon dioxide that can increase alkalinity of ground water. Other sources of alkalinity include run-off water over landscape, rich in limestone. Here, it is worth reiterating the difference between alkaline (directly related to pH) and alkalinity (measure of a solution's ability to neutralize acids). In the context of this work primarily dealing with alkalinity of irrigation water, the chief contributors are considered to be carbonate and bicarbonate ions. Even though hydroxyl ion (OH^-) contributes to total alkalinity, it is not expected to significantly affect the alkalinity of irrigation water. In other words, the term alkalinity in the entire manuscript refers to carbonate alkalinity (as opposed to total alkalinity). However, it is worth mentioning that the plasma-assisted approach can also be used to reduce hydroxide alkalinity. While pH and alkalinity values are extremely region-specific, it is worth mentioning some statistics for pH in various surface water stations in California. Out of the 28 California stations at which pH measurements are reported by USGS, 27 stations have a pH level higher than 7.0, with one station reporting a neutral pH.

A common approach to decreasing the pH as well as alkalinity to tolerable limits is acidification Enoch and Olesen (1993); Whipker et al. (1996); Albano et al. (2017) or the process of injecting acids into the irrigation water. The hydrogen ions in the acid will react with the bicarbonate/carbonate ions thereby producing carbon dioxide and water as given below



Candidate acids for lowering pH and alkalinity of irrigation water include sulphuric acid, nitric acid, and phosphoric acid with the specific choice depending on various factors including safety, cost, and potential side-effects to name a few. For example, nitric acid, inspite of possessing several advantages including addition of nitrate (NO_3^-) ions that assist plant growth is seldom used because of difficulties associated with handling concentrated nitric acid. It is worth mentioning that the addition of acid to water with bicarbonate/carbonate ions will lead to the formation of a corresponding salt (based on the acid and the cation attached to the bicarbonate ion) as a by-product but this has not been reported to be a major concern for plant growth. Magnetic treatment of irrigation water has also been considered as a candidate for soils with high alkalinity content Bogatin et al. (1999); Maheshwari and Grewal (2009). In this context, the use of electrical discharges ignited by the application of high voltage in ambient air promises to be an attractive alternative for on-the-fly acid generation and pH/alkalinity reduction. In fact, nature presents an excellent example of a similar process wherein lightning strikes lead to nitrogen fixation and enhanced soil fertility (albeit in limited quantities because of their short duration). The electrical discharges considered in this work can be classified as low-temperature laboratory plasmas in contrast to fully-ionized plasmas that are encountered in space as well as nuclear fusion. Low-temperature plasmas exhibit significant degrees of non-equilibrium characterized by hot electrons and cold ions/neutrals and are often referred to as non-thermal plasmas. The hot electrons of plasmas ignited in ambient air have sufficient energy to break the nitrogen and oxygen bonds and triggering a whole host of reactions leading to the formation of dissolved NO_3^- ions (amongst a whole host of other species) in the solution which, in

turn, can neutralize the bicarbonate ions. The interaction of atmospheric pressure plasmas with various biological substances has become an active area of research. This includes the use of atmospheric pressure non-thermal plasmas for water purification Malik et al. (2001); Malik (2010) and wastewater treatment Wang et al. (2012); Tyszler et al. (2006), surface modification for tissue engineering Chu et al. (2002); Chen and Su (2011); Intranuovo et al. (2014), agriculture Mitsugi et al. (2014); Ji et al. (2016); Li et al. (2016); Park et al. (2018), bacterial inactivation Traylor et al. (2011) and wound healing Bekeschus et al. (2016); Lee et al. (2016a,b) to name a few. Specifically, the role of plasmas in the area of water treatment has been growing exponentially with researchers studying the degradation of pharmaceutical compounds such as antibiotics Magureanu et al. (2015, 2011, 2010); Banaschik et al. (2015) and cyanide Hijosa-Valsero et al. (2013), enhancement of irrigation water Park et al. (2013); Ji et al. (2016); Sivachandiran and Khacef (2017); Judée et al. (2017) to name a few. The noticeable fact in previous representative work is the wide variety of plasma sources utilized to achieve the required effect. Plasma ignition has been pursued and demonstrated in the gas phase, in the liquid phase Foster et al. (2010) as well as multiphase with plasma formation in either gas phase with dispersed droplets or liquid phase with gas bubbles Sommers and Foster (2014). In spite of the large number of publications dealing with various aspects of plasma-liquid interaction Bruggeman et al. (2016), the interaction with alkaline water that contains dissolved bicarbonates/carbonates from the perspective of enriching irrigation water has not been considered in detail before. A robust energy-efficient technology for decreasing alkalinity in surface water would greatly benefit irrigation water quality thereby ensuring ideal growth conditions for the crop. An on-demand technique using limited raw materials such as the one proposed here would also ensure that there is no need to handle or transport acids utilized for neutralizing. In this context, the primary goal of the current work is to study the interaction of an atmospheric pressure electrical discharge set-up with varying concentrations of sodium bicarbonate dissolved in distilled water. Apart from decreasing the alkalinity by reacting with bicarbonate, the HNO_3 produced due to plasma treatment also assists in increasing the nitrate concentration thereby leading to an enrichment of the irrigation water. While the experimental study focuses on measuring pH and nitrate concentration as a function of plasma treatment time, the other objective of the current work includes the formulation of a system-level model that describes the treatment process thereby assisting in the design and scale-up of these plasma treatment systems. One aspect of the current work that is worth emphasizing is the use of only sodium bicarbonate to increase the alkalinity and pH of distilled water. In reality, the alkalinity in irrigation water has contributions from both carbonate and bicarbonate ions. This work chose to introduce only of these ions to increase alkalinity with the expectation of simplifying the experimental approach apart from reducing the design space (in terms of testing various bicarbonate to carbonate ratios). Therefore, the use of sodium bicarbonate (as opposed to sodium carbonate or a combination of both) was only by choice. However, it has been reported Somridhijev and Boyd (2016) that bicarbonate ion is the primary contributor to alkalinity under near-neutral pH (< 8.3) conditions and this fact motivated our choice. Also, it should be mentioned that the system-level model will remain the same even in the presence of sodium carbonate with the only difference being the initial concentration of carbonate ions.

The remainder of the manuscript is organized as follows. Section 2 describes the experimental set-up in detail with Section 3 describing the system-level model. Section 4 presents the results and discusses the same with Section 5 reserved for conclusions.

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