Experimental Studies on Physicochemical Parameters of Water Samples before and after Treatment with a Cold Atmospheric Plasma Jet and its Optical Characterization

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Cold plasma-liquid interaction becomes a growing interdisciplinary area of research involving plasma physics, fluid science, and chemistry. Plasma-liquid interaction has gained more interest over the last many years due to its potential applications in different fields. Cold atmospheric plasma jet is an emerging technology for surface drinking water treatment to improve quality and surface modification that is chemical-free and eco-friendly. Cold plasma treatment of water samples results in changes in turbidity, pH, and conductivity and in the formation of reactive oxygen and nitrogen species (RONs). As a result, plasma-activated water has a different chemical composition than water and can serve as an alternative technique for microbial disinfection. CAPJ has been generated by a high voltage 5 kV and a high frequency 19.56 kHz power supply. The discharge has been characterized by an optical method. To characterize the cold atmospheric pressure argon plasma jet, discharge plume temperature, and electron rotational and vibrational temperature have been determined. Cold atmospheric argon plasma jet produced at atmospheric condition contains high energetic electrons, ions, UV radiation, reactive oxygen, and nitrogen species named as cold plasma which has a wide range of applications in the biomedical industry, as well as in water treatment. Nowadays, researches have been carried out on ozonation through plasma jet interaction with surface drinking water. In this paper, we compare the change in physical and chemical parameters of surface water used for drinking purposes. The significant change in the physical parameters such as pH, turbidity, and electrical conductivity was studied. In addition, the significant changes in the concentration and absorbance of nitrate, ferrous, and chromium ions with respect to treatment time were studied. Our results showed that plasma jet interaction with surface drinking water samples can be useful for the improvement of water quality and an indicator for which reactive species play an important role in plasma sterilization.

1. Introduction

Plasma-liquid interaction has attracted increasing attention in recent years. The ability of plasma technology in plasma-liquid interaction, which has been confirmed due to the results of previous studies, along with the critical issue of water pollution as well as the lack of potable water, has led to the emergence of wide studies pointing to the use of different types of discharge plasma for water purification and wastewater treatment to improve their quality [1]. Non-thermal plasma is a type of plasma in which the electron temperature is much more than the ion temperature, but the overall plasma temperature remains low. Atmospheric plasma has been recognized as an important basic tool for...
promoting various chemical reactions at low temperatures. More specifically, the physical and chemical properties of plasma-treated water are outlined in relation to the acidity, conductivity, redox potential, and concentration of ROS and RNS [2, 3]. The simultaneous generation of physicochemical phenomena promotes itself to the top of the promising methods for liquid processing [4]. Cold atmospheric plasma jet (CAPJ) is a type of nonthermal plasma which has been used in various fields. The type and concentration of reactive species that are present in plasma-treated water depend on the gases and liquids used to generate plasma [5].

Plasma production in water affords one of the opportunities to thermally inject advanced oxidation processes into the water for chemical processing. Such technology could potentially change the treatment of drinking water [6]. Water activated by nonthermal plasma creates an acidified solution containing reactive oxygen and nitrogen species, known as plasma-activated water. The physicochemical properties of plasma-activated water stored at different temperatures were evaluated, including pH, conductivity, turbidity, nitrate, and nitrite anion concentration [7]. It has potentially a wide range of applications including environmental and biological fields such as food sterilization, cancer cell treatment, materials processing, and wastewater treatment. In package decontamination of foods, using cold plasma has advanced this technology as a single process for fresh foods decontamination and shelf-life extension [8, 9]. The cold plasma treatment is effective for both underground water as well as surface drinking water to enhance their quality. The degraded quality of water due to biological and chemical pollutants is improved by CAPJ treatment [10]. Besides that cold plasma also changes the concentration of physical parameters such as pH, turbidity, and electrical conductivity. Ozone generated from discharge and different highly reactive species formed in treated water are responsible to reduce the biological and chemical pollutants. In our experiment, we used CAPJ to compare some physicochemical parameters of water samples before and after treatment [11]. Similar results were reported by Hamdan et al. that the production of physicochemical process of interest promotes itself to the important promising technologies for liquid processing.

2. Materials and Methods

Figure 1(a) shows the schematic diagram of the experimental setup and Figure 1(b) shows the image of the discharge. The cold atmospheric plasma jet (CAPJ) system consists of a glass tube of diameter 3 mm through which argon is passed. Electrodes are separated by a distance of 5 cm. Flow of argon gas is maintained at the rate of 5 L/min. 10 ml of surface drinking water was treated for 5 and 10 minutes by a cold atmospheric plasma jet at an applied voltage of 5 kV and an operating frequency of 19.56 kHz. The length of the plasma jet was about 7 cm from the nozzle. Optical characterization of the discharge was done by calculating rotational and vibrational temperature using an optical emission spectrometer (USB2000+, Ocean Optics).

Figure 2(a) shows the photograph of Dudhpokhari tap (spring) water that is one of the biggest drinking water sources of the Kirtipur Municipality. Geologically, it lies on the Chandragiri limestone formation. It is located about 1390 m high from the sea level. Dudhpokhari spring might belong to the “seepage spring.” The seepage spring let a large volume of water out through loose soil or limestone rock. Figure 2(b) shows the CAPJ treatment to water samples collected from the tap water of Dudhpokhari, Kirtipur, near the capital city of Nepal. The treated samples of water were characterized using UV-visible spectrophotometer and the absorbance was recorded. The concentration of chemical species is observed using simple calibration curve. Produced cold plasma jet was injected for 5 to 10 minutes onto 10 ml water in a beaker. Before and after treatment, physical parameters such as pH, electrical conductivity, and turbidity were compared using pH meter, conductivity meter, and turbidity meter, respectively.

UV-visible spectroscopy is the measurement of the attenuation of a beam of light after it passes through a sample or after reflection through a sample surface. It measures the intensity of light passing through the sample (I) and compares it to the intensity of the light before it passes through the sample (I0). The ratio I/I0 is called the transmittance (%T) and the absorbance (A) is based on the transmittance which can be written as $A = –\log(\%T/100\%)$.

The basic parts of the spectrophotometer are the light source, a holder from the sample, a diffraction grating in monochromators, and a detector as shown in Figure 3.

A spectrophotometer can be a single beam as well as a double beam. In the single beam, all light passes through the sample. To be measured, the sample, whereas, in double beam, the light splits into two beams before it reaches the sample.

3. Results and Discussion

3.1. Temperature Measurement of Cold Atmospheric Plasma Discharge. Figure 4(b) shows the variation of temperature of plasma jet with time at discharge voltage 5 kV. It was linear up to 60 seconds and then almost remains constant at 25.5°C. Atmospheric plasmas have pressure approximately matching with surrounding atmosphere. This plasma is also called normal plasma. The plasma jet was designed with locally available materials to make treatment continuous and cost-effective. Temperature of plasma jet was directly measured as shown in Figure 4(a) which was about (24–27) 25.5°C at 5 kV, so it is called cold atmospheric plasma jet and widely used for comparing some physical and chemical parameters of surface drinking water samples before and after plasma jet treatment [12, 13].

3.2. Optical Characterization of CAPJ. The optical characterization of the discharge was carried out by using the line intensity ratio method. In this method, four suitable lines (two for Ar I and two for Ar II) were chosen from the
spectral lines of argon discharge. The working formula used to calculate the electron temperature is as follows [2, 14–16]:

\[
\frac{R_1}{R_2} = \left( \frac{I_1}{I_2} \right) = \left( \frac{I_{pq}}{I_{rs}} \right) \left( \frac{g_p}{g_q} \right) \left( \frac{\lambda_{rs}}{\lambda_{pq}} \right) \left( \frac{A_{pq}}{A_{rs}} \right) \left( \frac{g_u}{g_v} \right) \left( \frac{\lambda_{uv}}{\lambda_{xy}} \right) \exp \left[ \frac{-E_p - E_r - E_s + E_v}{K_B T_e} \right]
\]

Here, in equation (1), \( R \) is the ratio of the intensity of two lines, \( I \) is the intensity of the spectral line, \( A_{ij} \) is the transition probability of the transition \( i \rightarrow j \), \( g_i \) is the statistical weight of the upper level, \( \lambda \) is the wavelength of the line radiation, \( E_i \) is the energy of the upper level, \( K_B \) is Boltzmann constant, and \( T_e \) is the electron temperature. The
values of $\lambda$ and $I$ are obtained from the observation, and the values of $A_{ji}$, $g_j$, and $E_i$ are obtained from the National Institute of Standards and Technology (NIST) Atomic Spectra Database [17] (Figure 5).

Table 1 shows the different data of transition probability, statistical weight, energy of excited states, and the intensity at different corresponding wavelengths obtained from NIST database. By substituting these values in equation (1), the ratio of the intensities is expressed as

$$\frac{R_1}{R_2} = 1.14 \times 10^{-2} \exp \left( \frac{2.52}{K_B T_e} \right).$$  \hfill (2)

At different values of electron temperature, the values of the ratio of intensities of the corresponding wavelengths were determined. When the graph is plotted between the data of intensity ratio and electron temperature, the nature is as shown in Figure 6.

For the ratio of intensity of these four particular wavelengths, $((I_1/I_2)/(I_3/I_4)) = 1.14$. From the graph, the electron temperature ($T_e$) was found to be 0.58 eV. Similarly, the electron density was calculated by using the following equation [15]:

$$n_e = 2 \left( \frac{I_1}{I_2} \right) \left( \frac{A_1}{A_2} \right) \left( \frac{g_2}{g_1} \right) \left( \frac{2\pi m k T_e}{h^2} \right)^{3/2} \exp \left( \frac{-E_1 - E_2 + E_i}{k T_e} \right).$$  \hfill (3)

Substituting these parameters obtained from the National Institute of Standards and Technology (NIST) Atomic Spectra Database [17] in equation (3), the electron density was found to be $2.37 \times 10^{16}$ cm$^{-3}$ at 5 kV.

3.3. Determination of the Rotational Temperature of CAPJ.  The rotational temperature is essential, which is considered as the temperature of neutral particles present in plasma. The determination of the rotational temperature is the basis of Boltzmann law. The emission intensity of a specific spectral line is given by the formula expressed in the following equation [18]:

$$I_{em} = C (J' + J'') + 1 \exp \left( -\frac{B_j h c J'}{k_B T_{rot}} \right),$$  \hfill (4)

where $C$ is the constant for a specific band head, $J'$, $J''$ are the rotational quantum numbers for two transition states, $h$ is the Planck’s constant, $k_B$ is the Boltzmann constant, $T_{rot}$ is
the rotational temperature, and $c$ is the speed of light. Solving this equation, we have the following equation (5) to calculate the value of rotational temperature:

$$\ln\left(\frac{I_{on}}{J’ + J'' + 1}\right) = -\frac{B}{k_B T_{rot}} J’ (J'' + 1) + \ln C. \tag{5}$$

Equation (4) can be reduced to the form

$$T_{rot} = \frac{B}{k_B K}, \tag{6}$$

where $B$ is the rotational constant and $K$ is the slope of the linearly fitted graph presented in Figure 7. Putting the value of the slope of the straight line, Boltzmann constant, rotational constant, Planck constant, and velocity of light in the right hand side of equation (6), the rotational temperature of the plasma discharge is found to be about 295.97 K [18].

3.4. Determination of the Vibrational Temperature of CAPJ.

Emission intensity of a specific vibrational band of the spectral line is expressed in the following equation [19]:

$$I_{\nu’-\nu''} \propto h c v f_{\nu’-\nu''} A_{\nu’-\nu''} N(0) \exp\left(\frac{hcG(\nu’)}{k_B T_{vib}}\right). \tag{7}$$

where $I_{\nu’-\nu''}$ is the intensity of a vibrational transition, $f_{\nu’-\nu''}$ is the transition frequency, $C$ is the speed of light, $G(\nu’)$ is the vibrational term, $N(0)$ is the initial density of molecules, $h$ is

![Figure 5: Spectra of the discharge at frequency 19.56 kHz and an applied voltage of 5 kV in argon environment (flow rate = 5 L/min).](image)

![Figure 6: Graph of intensity ratio of the corresponding wavelengths as a function of electron temperature at 5 kV and an operating frequency of 19.56 kHz (argon flow rate 5 L/min).](image)
the Planck’s constant, $k_B$ is the Boltzmann constant, and $T_{vib}$ is the vibrational temperature.

Solving equation (7), we have found the following form to determine the vibrational temperature:

$$\ln \left( \frac{I_{y'\to y''}}{y'!} \right) = D - \frac{\hbar G(y')}{k_B T_{vib}}$$

(8)

Table 2 shows the vibrational transition and transitional probability at different wavelengths used for estimating the vibrational temperature. A graph is plotted between the data of intensity and wavelength, the nature is shown in Figure 8, and by the linear fitting method to determine the slope of the straight line.

Figure 9 shows the data points and their linear fit graph to calculate the slope of the straight line, and putting the values of different parameters in equation (8), the value of vibrational temperature is obtained to be about 2311.59 K [19].

3.5. Chemical Parameters

3.5.1. Ferrous Ion (Fe$^{+2}$). Figures 10 and 11 show the dependence of the absorbance and concentration of ferrous ions in the sample with respect to treatment time. To show the relationship between cold plasma treatment time with absorbance and concentration, light of wavelength 520 nm is used. Initially, absorbance and concentration of ferrous ions are 0.105 and 3.1 ppm. After treated 10 minutes, the absorbance and concentration of ions increase to 0.131 and 3.96 ppm, respectively, which are shown in the graph presented in Figure 10. The result shows that the concentration as well as absorbance gradually increased with treatment time. This is due to the reactive species generated during the process, which slightly increased ferrous (Fe$^{+2}$) ion concentration as well as its absorbance in the water samples after cold plasma treatment. The ferrous ions dehydrated with Y-zeolite are oxidized to ferric ions by the adsorption of dry oxygen. During the case, one oxygen atom absorbed more than two iron ions; therefore, the absorbance and concentration (linear as shown in Figure 11) of ferrous ions improved in the solution after plasma treatment [20].

3.5.2. Nitrite Ion (NO$^{2-}$). Figure 12 shows the dependence of the absorbance and concentration of nitrite ions in the sample on the cold plasma treatment time. To show the relationship between cold plasma treatment time with the absorbance and concentration, light of wavelength 520 nm is used. Initially, absorbance and concentration of nitrite ions are 0.105 and 45 ppm. After treated 10 minutes, the absorbance and concentration of ions increase to 0.18 and 82 ppm, respectively, which are shown in the graph presented in Figure 12.

Figure 13 shows the relationship between concentration and absorbance; these parameters increase with treatment time, which is shown in Figure 12. This is due to the fact that ozone (O$_3$) and nitrogen oxide (NO$_3^-$) generated during the discharge in air convert nitrite or other nitrogenous compounds in the water into nitrate by direct oxidation. Therefore, the concentration of nitrite ions increased in the treated water samples which was the cause of the absorbance peak at wavelength 520 nm [21, 22].

3.5.3. Chromium Ion (Cr). Chromium (Cr) occurs naturally in the environment and has potential risks for human beings. Cr exists in many oxidation states in environment. Chromium ions are an essential nutrient for maintaining liquid insulin and glucose metabolism and its scarcity leads to diabetes. Figures 14 and 15 show the dependence of the absorbance and concentration of chromium ions in the water samples after the plasma treatment. To show the relationship between cold plasma treatment time with absorbance and concentration (absorbance versus concentration is linear as shown in Figure 15), light of wavelength 440 nm is used. Initially, absorbance and concentration of chromium ions are 0.075 and 92 ppm. After treated by plasma for 5 minutes, the absorbance and
Table 2: Vibrational transition and transitional probability at different wavelengths.

| Vibrational transition | Wavelength $\lambda$ (nm) | Transitional probability $(v' - v'') S^{-1}$ |
|------------------------|-----------------------------|-----------------------------------------------|
| 1–0                    | 315.801                     | $1.266 \times 10^6$                          |
| 1–2                    | 353.556                     | $5.590 \times 10^6$                          |
| 0–1                    | 357.578                     | $8.905 \times 10^6$                          |
| 2–4                    | 370.933                     | $4.095 \times 10^6$                          |
| 0–2                    | 380.378                     | $3.532 \times 10^6$                          |
| 2–5                    | 394.172                     | $3.081 \times 10^6$                          |
| 1–4                    | 399.708                     | $2.375 \times 10^6$                          |
| 1–3                    | 375.22                      | $4.885 \times 10^6$                          |

![N$_2$ (SPS) Experimental vibrational spectrum](image1)

**Figure 8:** N$_2$ (SPS) vibrational spectrum and experimental curve fitting.

![Experimental curve fitting](image2)

**Figure 9:** Graph to determine the vibrational temperature of CAPJ (5 kV, 19.56 kHz, and 5 L/min).
concentration of chromium ions increased linearly up to 0.15 and 190 ppm, and then after treated for 10 minutes, the absorbance and concentration of ions gradually reduced to 0.12 and 150 ppm, respectively, which are shown in Figure 14.

The result in Figure 15 shows the concentration as well as absorbance at wavelength 440 nm after treatment, first increasing and then gradually decreasing but linear to each other at a particular wavelength. Finally, the produced reactive species (RONS) in the plasma discharge during the process is efficient in the addition of chromium ions to the samples after treatment. So, finally after 10 minute treatment to water samples, chromium ions found slightly increased [23].

3.6. Physical Parameters

3.6.1. Conductivity. Electrical conductivity is a measure of water ability to convey electrical current, which is related to the concentration of ions present in water. The experiment shows that the conductivity of the water sample increases with time. The conductivities of the untreated and argon plasma jet treated samples collected from drinking tap water are presented in Figure 16. The conductivity of the untreated sample was found to be 290 μS/cm. After treatment by a cold atmospheric plasma jet for 5 and 10 minutes, its conductivity slightly increased to 317 μS/cm and 320 μS/cm, respectively. The mean value of the four times collected samples before and after treatment was presented as a histogram in Figure 16.

To change in the conductivity of water, the conductive ions can come from dissolved salts and inorganic materials such as chlorides, alkali, and carbonate compounds [24]. After treatment by a cold atmospheric argon plasma jet, the electrical conductivity was found to be slightly increased. There is a slight change in conductivity before and after treatment. This result has good agreement with the results presented by Lawaj et al. [25].
3.6.2. pH. pH is expressed as the intensity of the alkaline or acid condition of a solution. Most of the waters are alkaline in nature due to the presence of carbonates and bicarbonates. Generally, the pH value of water is in the range of 6.5 to 8.5. This range of pH of water is recommended for the domestic purpose [26]. pH refers the concentration of hydrogen ions within the water samples. Initially, the pH value of a four time collected untreated sample of water was found to be 8.2. After treated by cold argon plasma jet for 5 minutes and 10 minutes, its value decreased to 7.5 and 7.4, respectively. It means water became more neutral (pH about 7.5) after the plasma treatment. This indicates that there is a slight decrease in the pH value after treatment. Cold plasma contains reactive species such as nitrogen oxide (NO), ozone (O₃), and hydrogen peroxide (H₂O₂) ions. After treatment, ozone oxidizes the free ions in the solution. The pH levels hence slightly decreased in argon discharge due to acidification, which is shown in Figure 17. The pH decrease in water can be attributed due to the presence of reactive species such as hydrogen peroxide and nitrogen oxide. This result is correlated to the result presented by Niguette et al. [27, 28].

3.6.3. Turbidity. Turbidity is the cloudiness of a fluid caused by the maximum number of particles which are invisible to our naked eye, similar to that of smoke in air mixture. Especially, the fluid consists of suspended impurities due to inorganic matter or microorganism of different sizes [29]. Initially, the mean value of turbidity of four times collected surface drinking water samples was found to be 3.8 NTU. After cold argon plasma jet treatment for 5 minutes and 10 minutes, the values of turbidity were reduced to 2.9 NTU and 2.22 NTU, respectively. The turbidity has decreased as per the treatment time that represents the cold plasma has efficacy in destroying microbes or pathogens and other agents present in water samples. There is reduction in turbidity after plasma treatment, which is shown in Figure 18. After treatment, the turbidity is effectively reduced that indicates the impurities present in the samples were

![Figure 14: Absorbance and concentration of chromium ions with treatment time (5 kV, 19.56 kHz, and 5 L/min).](image1)

![Figure 15: Calibration curve (linear fitting) for determination of chromium ions.](image2)
Figure 16: Variation of conductivity with treatment time (5 kV, 19.56 kHz, and 5 L/min).

Figure 17: Variation of pH with treatment time (5 kV, 19.56 kHz, and 5 L/min).

Figure 18: Variation of turbidity with treatment time (5 kV, 19.56 kHz, and 5 L/min).
killed by reactive species produced in the plasma discharge. Ozone generated during the plasma discharge is directly affecting the microorganism which is responsible for destroying pathogens in the treated samples. This result is correlated with the result presented by Dyas et al. [29–31].

4. Conclusion

A cost-effective system of cold plasma jets generating at atmospheric conditions with potential application in plasma-liquid interaction has been developed. Temperature of the plasma jet was directly measured about 25.5°C, so the produced discharge is termed as cold plasma and is widely used in comparing the physical and chemical parameters of drinking water before and after treatment. Cold atmospheric plasma jet has been characterized by an optical method. The electron temperature and electron density of the CAPJ were found to be $0.58 \text{ eV}$ and $2.37 \times 10^{16} \text{cm}^{-3}$, respectively, at 5 kV using intensity ratio method. The rotational and vibrational temperatures of the discharge were found to be 295.97 K and 2311.59 K, respectively. The reactive species produced in the cold atmospheric plasma jet affected physical and chemical parameters of water samples after treatment. Experiments showed that the linear variation of concentration and absorbance of nitrite, ferrous, and chromium ions in water at wavelengths 520 nm and 440 nm were found to be increased after treatment. -Y_-His approach was partially supported by the Nepal Academy of Science and Technology (NAST), Nepal, for providing PhD Fellowship through Grant no. 11/073/074. The authors would like to acknowledge Dr. Bhagirath Ghimire for calculating rotational and vibrational temperatures. The authors would also like to acknowledge Department of Physics, Kathmandu University, Institute of Science and Technology (IOST), Tribhuvan University, Nepal, for their invaluable help and support.

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