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Degradation of Telmisartan (TELMA-H) in Simulated Aqueous Solution by Non-Thermal Atmospheric Pressure Plasma Jet (APPJ)

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ABSTRACT

In this study we investigate the degradation of telmisartan (Telma-H) in simulated aqueous solution using non-thermal atmospheric pressure plasma jet (APPJ). Aqueous solution containing Telma-H was treated with APPJ as a function of applied potential and reaction time. The degradation of Telma-H was investigated by means of UVVisible spectroscopy. Optical emission spectra (OES) of the plasma jet was used to identify the reactive species that contributed to degrade Telma H compounds. The variation of pH and conductivity of the plasma treated Telma H aqueous solution was also measured.

KEYWORDS

T-H wastewater; Degradation; OES; APPJ; EC.

1. Introduction

Pollution of water resources and aquatic environments like river, pond, and ground water by organic compounds and pharmaceuticals is a serious environmental problem in recent years. The presence of pharmaceutical and medical wastes in the green environment is a matter of serious concern as it affects living organisms and ecosystems [1]. The pharmaceutical compounds pollute water sources via animal and human medical waste containing drug, large amount emission from pharmaceutical production sites, direct disposal of unused medicine by hospitals and human beings. Furthermore, the use of these pharmaceutical related products cannot be reduced or eliminated due to the increasing population growth. Most of the pharmaceutical compounds are non-biodegradable and stable in aquatic environment. Concurrently, many pharmaceutical compounds are harmful even in very low concentration [23] and therefore, it is necessary to develop efficient water treatment process for reducing or eliminating pharmaceutical wastes in aquatic environments. Conventional methods like filtration, sedimentation, biodegradation are ineffective to remove pharmaceutical compounds and organic compounds. Advanced oxidation process (AOPs) can be used to for degradation and elimination of pharmaceutical wastes efficiently. AOPs such as non-thermal atmospheric pressure plasma jet (APPJ) process are very effective and efficient alternative to conventional methods. Since last decade, the APPJ treatment process has received considerable attention for removal of pharmaceuticals and toxic organic compounds from water.

Furthermore, in APPJ-assisted AOP high energetic nitrogen, oxygen species, and OH radicals are generated, which destroy and oxidize the organic molecules. The APPJ assisted process is eco-friendly and ensures complete destruction and removal of organic compounds [4-5]. The pharmaceutical compound T-H tablet (medicinal compound containing Telmisartan and Hydrochlorothiazide) is widely used for treatment of high blood pressure, kidney failure, and liver failure [6]. The aim of the present work is to study the degradation of commercially available T-H tablet using nonthermal APPJ. The degradation of the T-H tablet was investigated with respect to various plasma parameters such as applied potential and treatment times. Degradation of the T-H tablet in aqueous solution was carried out and the degradation products were characterized using various analytical techniques.

2. Experimental Procedure

Telma-H tablet was used for the studies. A stock solution of T-H drug in deionized water containing 10-4 mole concentration of the drug was prepared and samples from this stock solution were used for experiments. Samples of the aqueous solution of the drug were treated by APPJ at different applied potentials (29, 31 and 33 kV) and treatment time (05, 10, 15, 20 and 25 min).

2.1 Non-Thermal Atmospheric Pressure Plasma Jet (APPJ)

The atmospheric pressure plasma jet (APPJ) device consists of a copper rod and copper ring, which act as the live electrode(cathode) and ground electrode respectively.

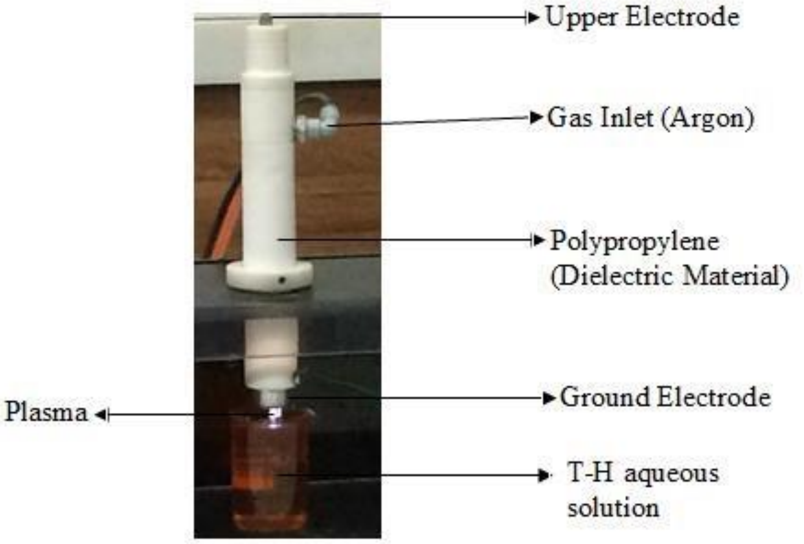

Figure 1. Photographic image of Non-thermal APPJ

2.2 Analysis of the Plasma Treated Samples

Aqueous samples of T-H pharmaceutical solution were analysed by various analytical tools after exposure to APPJ. The concentration of the T-H drug was measured spectrophotometric ally (Ocean Optics HR 4000) after plasma exposure. The electrical conductivity of the samples was measured using an Elico digital electrical conductivity meter-611 (Eloco Ltd, India) and pH of the water samples was measured using HANNA pH meter.

3. Results and Discussion

3.1 Optical Emission Spectra Analysis of RONS and Argon Emission

Fig 2a and 2b show the optical emission spectra of the APPJ (without the sample) and the plasma jet during plasma treatment respectively. The Ar-plasma jet was generated at 29 kV and it exhibits well-defined argon emission peaks in the wavelength range of 650-900 nm. Spectral peak of weaker intensity, attributed to OH-radicals

Was also observed at 309 nm. The OH radicals are produced as a consequence of the plasma jet interacting with water molecules and oxygen in the surrounding atmospheric. The intensities of the spectral peaks of OH radicals and argon emission lines were found to increase with increasing applied potential and this may be caused by higher energy electrons and excited atoms produced at higher potential.

The generation of reactive oxygen and nitrogen (RONS) species was investigated by OES (Fig 2b) during plasma treatment. The most intense argon emission lines can be found in the wavelength region of 680-850 nm. The plasma jet also produced a significant amount of OH radicals, first and second positive species of N2 and atomic O2 at 309 nm, 320-400 nm ,774 nm, and 842 nm respectively [8-9]. These emission spectral lines result by interaction of the plasma jet with the drug sample solution and water vapour and air in the surrounding atmosphere and consequent formation of OH radicals and reactive nitrogen species. It was also observed that the concentration of RONS species and hence the spectral intensity increased with increasing applied potential. Subsequently, these reactive species interact with water molecules to ionize and oxidize the pharmaceutical drug molecules to nontoxic simpler species. Therefore, the RONS species play an important role in the degradation of T-H molecules.

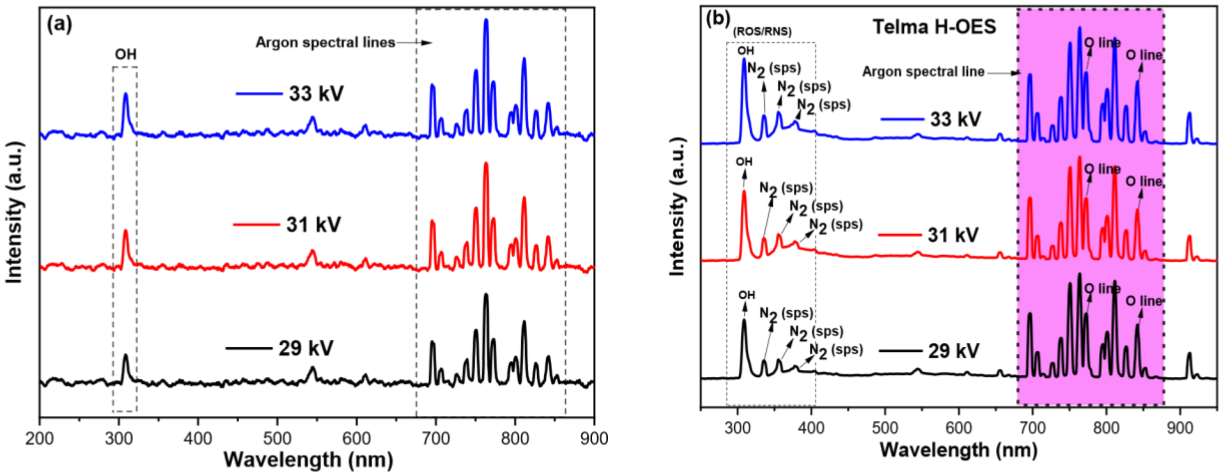

Figure 2. The optical emission spectrum of argon spectral lines with and without T-H aqueous solution

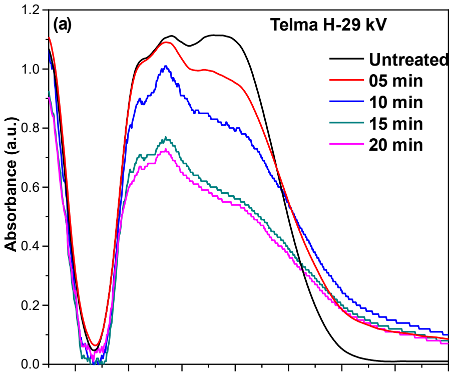

Figure 3. The UV-visible absorption spectrum of Telma-H

3.2 UV-Visible Absorption Analysis also as the Discharge Potential Is Increased. The Degradation of the T-H Drug by Plasma Treatment into Simpler Organic

The T-H drug concentration in the aqueous solution species is evident from the above results. The highest peak was measured by UV-Visible absorption spectroscopy after intensity of T-H was observed at 274 and 293 nm [6]. The plasma exposure at different discharge potentials and intensity of these peaks decreased with increasing plasma reaction time. The results are depicted in Fig 3, which clearly exposure time at 29 kV of discharge potential due to shows that the T-H concentration decreases after exposure to degradation of the drug molecules; the intensity of plasma. It is further found that the decrease in drug absorbance peak is a direct indicator of the presence of T-H concentration is more as the exposure time increases and molecules in the aqueous solution.

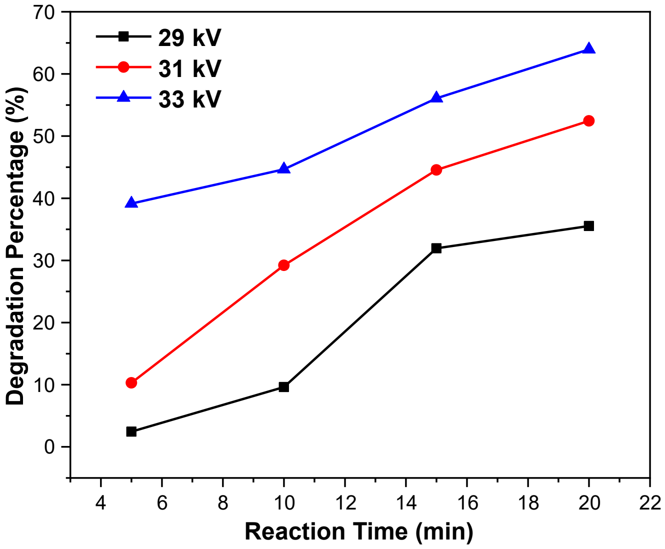

Figure 4. T-H drug degradation efficiency spectrum

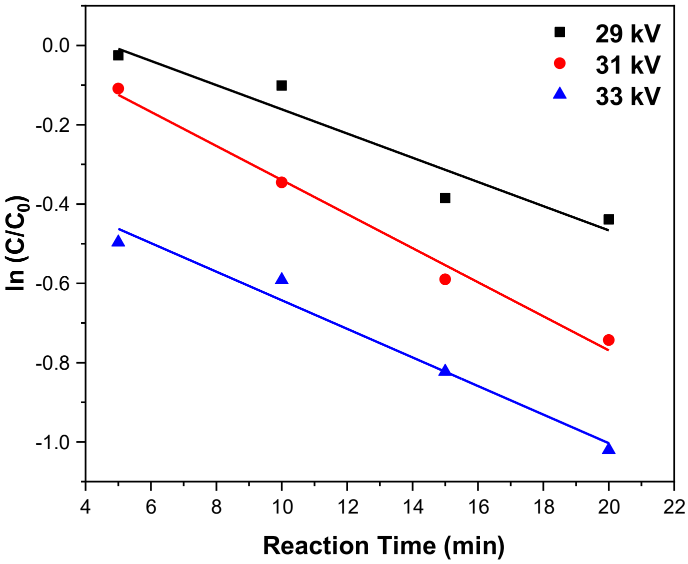

Figure 5. First order kinetic equation of T-H drug degradation

Furthermore, the degradation rate of 63% was obtained at 33 kV applied potential and plasma exposure time of 20 min.

Fig.4 shows the % of degradation of T-H drug as a function of plasma exposure time and discharge potential as calculated using the following expression

C0 − Cat% of degardation = × 100% C0

Where, η is the degradation % of the T-H drug, C0 and Cat are the initial and final concentration of the T-H drugs in the sample before and after plasma treatment.

3.3 Kinetic Equation of T-H Aqueous Solution Degradation

The non-thermal plasma assisted degradation of commercially available T-H as a function of discharge potential and reaction time are shown in Fig 5. T-H degradation was carried out under various plasma parameters. The degradation of T-H was observed to follow a pseudo-first-order kinetics [10-11]. The following expression is used to calculate the degradation rate constant.

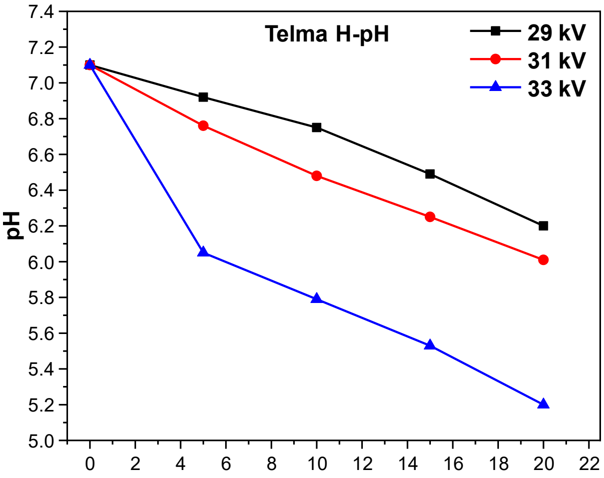

Figure 6. Variation of H of plasma treated T-H solution as a function of various operating parameter

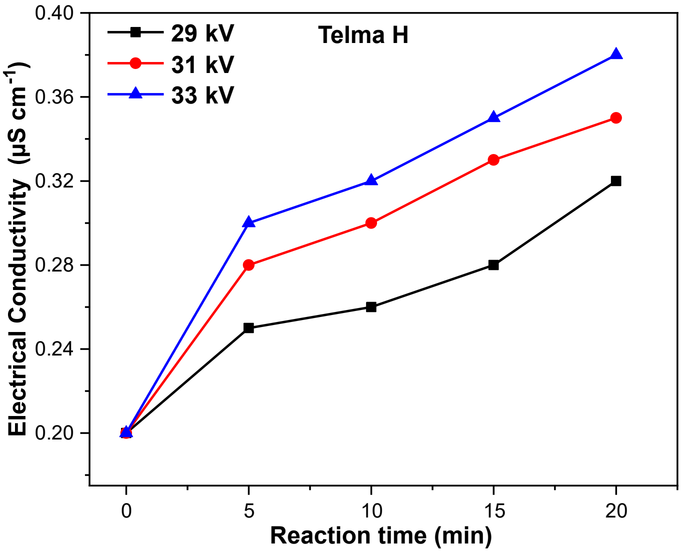

Figure 7. APPJ treated T-H aqueous solution Electrical conductivity graph

3.4 Temporal Change of pH in T-H Aqueous Solution C Temporal Value of pH of the Aqueous T-H Drug

Where, k is the first order rate constant, t is the plasma are shown in Fig 6. Fig.6 clearly shows that the pH of the treatment time (min) and C and C0 are the T-H. solution decreased with treatment time and applied potential 11], indicating that an acid is formed as one of the degraded Concentration before and after plasma treatment. It is seen from Fig.5 that the extent of degradation increases products of T-H drug. It is observed from Fig.6 that the pH with the discharge potential. Further, at a given discharge value decreased significantly (initial pH 7) with increasing potential, the degradation rate is found to increase with applied potential. Furthermore, the aqueous solution plasmaplasma exposure time. Among the studies samples, treated at the higher applied potential of 33 kV had the maximum degradation (63%) of the drug was observed after 20 minutes exposure at the plasma discharge potential of 33 lowest pH value of 4.8 obtained (Fig.6). The decrease in pH kV. The increase in degradation of the dye at increased value after plasma exposure clearly shows formation of an discharge potential can be attributed to higher concentration acid as one of the degraded products of T-H drug. Formation and higher energy of the reactive species. of reactive nitrogen species (as observed in the OES)

3.5 Electrical Conductivity Measurement of APPJ Treated T-H Aqueous Solution

Electrical conductivity (EC) is frequently used as an index to measure water quality as conductivity is directly related to the ion concentration in the aqueous solution [12]. The electrical conductivity of plasma-treated T-H aqueous solution is shown in Fig.7, which shows that the conductivity increases with treatment time and discharge potential. The variation in electrical conductivity with the processing parameters is similar to the variation of pH with the processing parameters. The increase in conductivity is due to an increase in the concentration of ionic species which are formed as degradation products of the drug. The results are in agreement with the observation that the pH value of the drug solution decreased with plasma exposure due to formation of acid species. The increase in conductivity of the drug solution after plasma exposure can be attributed to the presence of H+ ions (of the acid) produced as a result of fragmentation and degradation of the T-H drug.

4. Conclusion

Degradation of T-H drug solution by non-thermal atmospheric pressure plasma jet is presented in this paper. Plasma degradation of T-H aqueous solution was carried out under different applied potentials and plasma treatment times. Extent of degradation of the drug was followed by UVVisible spectroscopic studies. Higher degradation rate of T-H was observed at higher applied potential and treatment time. It was found that plasma degradation followed pseudo first order kinetics. Reactive OH radicals were found to be the species responsible for degradation of the drug. This was confirmed by optical emission spectra recorded during plasma treatment of T-H drug solution. Electrical conductivity and pH measurements of the drug solution after plasma treatment confirmed the formation of acid molecules as a fragmented compound from the T-H drug. Finally, the above results showed that the non-thermal APPJ treatment process is efficient and eco-friendly process that destroys the T-H drug forming simpler molecules.

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