Investigation of the usage of plasma systems in the field of environmental engineering

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ABSTRACT
In the study; It is planned to design a plasma jet reactor that allows liquid (water) purification at atmospheric pressure for the first time in our country, and to investigate to what extent it will contribute to the treatment. The study is important in terms of investigating the usability of plasma systems, which is an innovative technology, in the field of environmental engineering and contributing to the literature. In the light of literature research, it is observed that studies in the field of plasma systems and environmental engineering are generally aimed at color removal. In the literature, the removal efficiencies of Methylene Blue (MB), Methyl Orange (MO), Congo Red (KK)) are mostly compared as model dyes. Argon feed gas is generally used in the studies. When the direct treatment results are evaluated for three different dyes at 20 and 30 min treatment times, it is observed that high removal efficiencies such as MB 95%, MO 97%, KK 86% are obtained as a result of 20 min treatment. As a result; In our study, the usability of the atmospheric plasma system, which is a new technology in the field of water treatment in our country, was evaluated, inspired by the studies on the usability of plasma technology in water treatment in the world.

Keywords: Plasma Systems, Methylene Blue, Plasma Treatment, Atmospheric Pressure Plasma.

1 INTRODUCTION

The use of plasma technology in the field of environmental technologies is gaining importance in our country. From an environmental point of view, the usability of plasma systems in many areas such as treatment of drinking water and wastewater, disposal of solid wastes, modification of membrane surfaces used in wastewater treatment, etc. is being investigated.

The first scientific studies on plasma in history started by Humphry Davy. In 1874, William Crookes described plasma as the fourth state of matter. Systematic studies of plasma began in the 1920s with Irving Langmuir and other researchers. The term 'plasma' was first used by Irving Langmuir in 1928 as a description of ionised gas. Plasma, which is the fourth state of matter, is defined as an ionised gas mixture, which is separated into freely moving positively charged ions and negatively charged electrons by the ionisation of molecular atoms when the uncharged gas is exposed to sufficient energy.

99 percent of the universe is in plasma. Plasmas are characterised by a mixture of electrons, ions and neutral particles and are electrically neutral on average (i.e. the total charge of a plasma is roughly zero) and electrically conductive due to the presence of free charge carriers. To produce plasma at low pressure, an electrical voltage of 1000-2000 V is applied, while high voltages such as 20-40 kV are applied at high pressures. Plasma is different from other states of matter. It is not correct to call every ionised gas a plasma. Plasma is a good electrical conductor and generates an electromagnetic field. Any propagation in the plasma takes place at the speed of an electromagnetic wave. In the plasma state, chemical reactions take place at greater speeds. Coulomb attraction forces between charged particles in plasma are effective even at very long distances. Therefore, every particle in the plasma is in constant interaction with all particles around it. Gases do not conduct electricity and do not create electromagnetic fields. Any propagation in the gas takes place at the speed of sound. In the gaseous state, chemical reactions take place at slower speeds, and the Coulomb interaction between particles is only as far as the sum of two particle radii. Apart from these distances, particle interaction is ignored. Plasma can be formed naturally or under laboratory conditions. Plasma is classified in different ways according to the cause of its formation and its properties.

Table 1. Classification of plasma according to temperature

NATURAL PLASMAS		**ARTIFICIAL PLASMAS**	
Hot Plasmas	**Cold Plasmas**	**High Temperature Plasmas**	**Cold Plasmas**
Sun	Aurora	Thermonuclear	Arc Plasma
Stars	Sun	And Laser Fusion	Plasma
Lightning			Torch
	Wind		APPJ
			DBD

Source: Prepared by the authors.

Table 2. Classification of plasma according to hardware properties.

GLOW DISCHARGE	**ELECTRICAL BARRIER DISCHARGE**	**CORONA**	**DISCHARGE**
Low Frequency (AC)			
Radiofrequency (RF)			
Microwave (MW)			
Pulsed (DC)			

Source: Prepared by the authors.

Table 3. Classification of plasma according to pressure.

Atmospheric Pressurised Plasma (High Pressure - 1 Atm)	Vacuum Plasma (Low Pressure <1 Torr)

Source: Prepared by the authors.

Plasmas are an innovative technology and are used in many fields and their usage area continues to increase. These are; Plasma water treatment systems in the environmental field, aircraft and UAVs, fuel cells, cutting / welding machines, surface technologies, sterilisation (food and medical use), combustion systems, polymerisation and coating, space technologies, electronic chip making, hazardous and harmful waste (nuclear, garbage and medical waste) treatment.

Since thermal plasmas enable the degradation of all kinds of harmful components in reactions such as gasification, incineration and waste disposal thanks to their high temperature and make it possible to significantly reduce the rate of harmful emissions, the interest in plasma in this field increases considerably in the world. The fact that plasma can reach high temperatures and provide an efficient environment for chemical reactions with the charged particles it contains brings its use in the field of energy to the forefront.

There is a dramatic increase in the amount of organic pollutants (volatile organic compounds, drugs, organic dyes ...) detected in water resources day by day and this situation poses a threat to public health. The removal of colour from textile industry wastewater represents an important environmental objective as it allows water reuse. However, paints are designed to resist degradation and therefore degradation is not an easy task (García et al., 2017).

A large group of organic micropollutants are dyes used commercially in textile industries. Statistically, about 20% of industrial water pollution is caused by the discharge of dye wastes. It is one of the largest contributors to water pollution. Consequently, some dyes are responsible for catastrophic damage to aquatic ecological systems, attributed to their stability in nature. In addition, recent studies show that some dyes are carcinogenic (Kumar, Skoro, Gernjak, & Puac, 2021). Today, atmospheric pressurised plasmas are recognised as a very promising technology and studies are proving the efficiency of these systems. Conventional water treatment methods are ineffective in the degradation of most organic pollutants. Atmospheric pressurised systems are a highly effective method to break down these pollutants. Plasma systems will contribute to the applications in the field of green chemistry in recent years as an alternative to reduce or eliminate the use of chemicals used in advanced oxidation processes. In particular, it creates less toxic conversion intermediates that can be biodegraded during processing. It is also noteworthy that cold atmospheric pressure systems can be widely used in applications requiring low temperature.

In this study, inspired by the use of plasma technology in water treatment in the world, the usability of atmospheric plasma system, which is a new technology in the field of water treatment in our country, is evaluated.

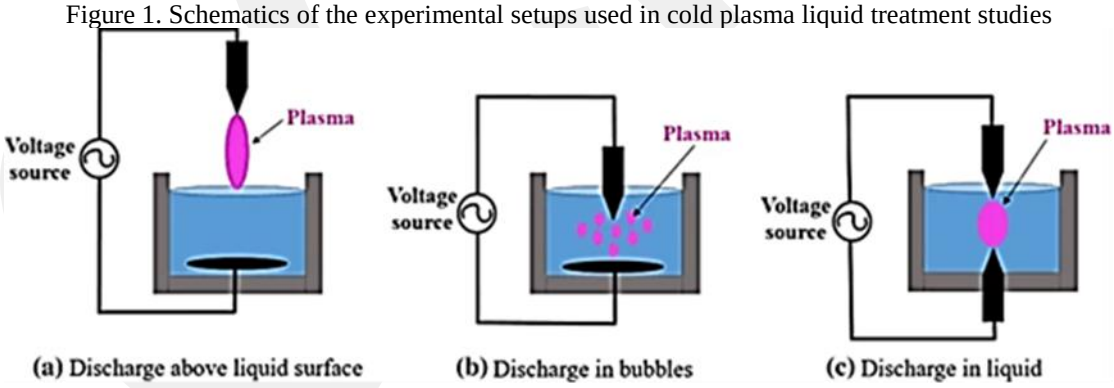
2 MATERIAL METHOD

In recent studies, the effectiveness of the system is tested during the degradation of the target pollutant model paint by atmospheric pressurised plasma jet.

2.1 INTERACTION OF PLASMA WITH LIQUID

In general, cold atmospheric plasma setups used for liquid interactions are divided into three categories: discharge over the liquid surface, discharge in bubbles, and direct discharge into the liquid, as shown in Figure 1. Kumar et al. (2021) mention plasma-liquid interactions in their study, 'Discharge over the liquid surface and in bubbles are widely used for wastewater treatment, while direct discharge into the liquid is not often used. In the case of a discharge above the liquid surface, plasma species are generated in the gas phase near the liquid surface and then transferred into the liquid sample for degradation of organic pollutants in the water.

Figure 1. Schematics of the experimental setups used in cold plasma liquid treatment studies



Source: (Kumar et al., 2021).

2.2 SAMPLE PREPARATION PROCEDURES

Chandana et al. (2015) worked with different concentrations of Methylene Blue in their research and observed that the degradation efficiency decreased with increasing methylene blue concentration, probably due to the presence of a higher number of reactant molecules. Using argon plasma jetting, they

achieved approximately 100% degradation efficiency for 20 ppm methylene blue. These rates decreased to 99%, 97% and 96% for 30, 40 and 50 ppm.

2.3 GAS FLOW RATE

Rahimpour et al. (2019) added additional Argon gas to the system in a study they conducted. By adding 90 ml/min Argon gas to the oxygen inlet, they observed an increase of approximately 44% in decolourisation at a flow rate of 90 ml/min. The addition of argon as an inert gas caused an increase in Crystal Violet decolourisation compared to pure oxygen. It was concluded that the higher the Ar flow, the faster the degradation rate. It was also proved that reactive species are produced in the gas phase during the discharge process. These species were then transferred to the liquid phase and reacted with contaminant molecules.

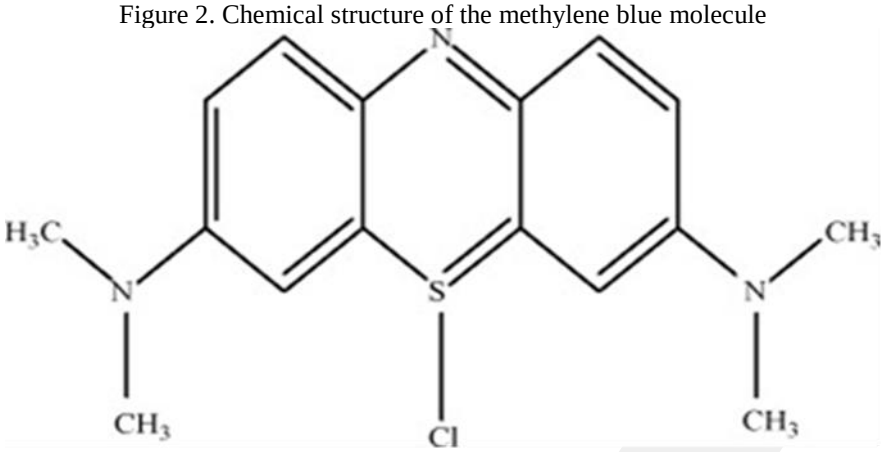
2.4 CHEMICAL ADDITION

Chandana et al. (2015) improved the performance of the system by adding iron as a catalyst, thus enabling the formation of the Fenton reaction, which mineralises methylene blue and leads to the formation of hydroxyl radical, a strong oxidant.

2.5 METHYLENE BLUE REMOVAL STUDIES

Huang et al. (2010) selected methylene blue as a model organic pollutant due to its stable molecular structure. They explained this with the theory that 'the lower the bond dissociation energy in chemical reactions, the more active the chemical bond is and the easier it is for old bonds to break and new bonds to form'. Accordingly, it is known that methylene blue molecule is easier to decompose in the presence of radical species and chemical bonds are easier to break and oxidised until they turn into inorganic ions. In the final stage of the plasma process, many radical species are produced in the methylene blue solution. These oxidise organic molecular structures to inorganic ions such as, $\boldsymbol{CO_2}$, $\boldsymbol{H_2O}$, $\boldsymbol{Cl}$, $\boldsymbol{SO_4^{2-}}$ve $\boldsymbol{NO^{3-}}$. The concentration of methylene blue solution is quantitatively analysed by spectrophotometric methods. Figure 2 shows the chemical bond structure of methylene blue.

Figure 2. Chemical structure of the methylene blue molecule



Source:(Huang et al.,2010)

3 RESULTS

In some recent studies on treatment, the reasons for the preference of atmospheric pressure plasma systems are investigated. Açıkel (2018) mentions the advantages of atmospheric pressure systems as follows: It has various advantages such as not requiring water, the process takes place in the gas phase, minimising the rate of chemical use, not causing industrial waste and making effective modifications without losing the basic properties of the material. Güleç (2011) in one of his studies mentioned the most important features of atmospheric pressure plasmas between atmospheric pressure plasmas and low pressure plasmas as follows: 'It is an important feature that atmospheric pressure systems do not contain expensive and complex components such as vacuum systems, vacuum control units, pressure gauges, gaskets and fittings in low pressure plasma systems. The variety of gases that can be used in this type of plasma is high and this increases the variety of particles suitable for chemical reactions in the plasma. In addition, the fact that the plasma obtained is not at high temperatures also expands the application area. For a plasma source to be created at atmospheric pressure, it is important that it is regular, adaptable to different applications and usable in the industrial field. Dojčinović, et al. (2011) in their study; 'Various conventional methods such as adsorption, coagulation, filtration, sedimentation and biological treatments have been used to treat paint wastewater, but none of these treatments have been satisfactory due to the high degree of polarity and complex molecular structure of the wastewater. Furthermore, they all result in producing a secondary waste product that needs to be further addressed.'' The problems of conventional methods are mentioned. Atmospheric pressure plasma systems effectively break down organic pollutants that are difficult to break down, such as paint. In addition, these systems are a very environmentally friendly system with no secondary waste. In a study by Kumar et al. (2021) in terms of energy consumption, the cold plasma system requires less energy to produce reactive species than the thermal plasma system. We can consider plasma decontamination processes as an environmentally friendly

technology. Yamada et al. (2020) in their study; 'Among many plasma production methods, low-temperature, atmospheric pressure discharge plasma is stable and easy to use. In summary, atmospheric pressure plasmas, which are an innovative system today, are considered a very promising technology and the efficiency of these systems is proven in line with the studies carried out. Traditional water treatment methods are ineffective in the degradation of most organic pollutants. Atmospheric pressurised systems can be preferred as a highly effective method to break down these pollutants.

In recent years, studies on the effectiveness of plasma systems in water treatment have increased. Table 4 and Table 5 summarise some of the recent studies on the subject.

Table 4. Comparison of some studies on plasma systems for organic dyes (model dye, removal method, feed gases, added chemicals, pH, conductivity)

Referances	**Model Dye**	**Removal Method**	**Feed Gases**	**Added Chemical**	**pH**	**Conductiviy (µS/cm)**
(Jiang, Zheng, Liu, & Wu, 2012)	Methyl Orange	Circulating Airtight Non-Thermal Plasma Reactor	Oxygen	Hydrochloric Acid, Sodium Hydroxide, Sodium Chloride	-	-
(Chen et al., 2018)	Crystal Violet	Dielectric Barrier Discharge	Air	Ferrous Ion, Sodium Persulfate	3-7	-
(Chen et al., 2017)	Crystal Violet	Dielectric Barrier Discharge	Air	-	3,55-2,93	102-203
(Rashid, Chowdhury, & Talukder, 2020)	Remazol Blue, Remazol Red, Remazol Yellow	Underwater Parallel Multi-Tube Air Discharge Plasma Jet	Air	-	25 min. RB:7,45-7,15 RR:7,05-6,55 RY:7,08-6,65	25 min. RB:104,5-112 RR:109,5-125 RY:106-116
(Chandana, Reddy, & Subrahmanyam, 2015)	Methylene Blue	Atmospheric Pressure Plasma Jet	Air, Nitrogen, Argon	Iron, Titanium Sulfate	Argon=3.2 Nitrogen=3,8 Air=2.8	Argon=700 Nitrogen=1600 Air=1700
(Abdel-Fattah, 2019)	Methylene Blue	Atmospheric Pressure Plasma Jet	Helium	-	-	-
(Yamada, Wahyudiono, Machmudah, Kanda, & Goto, 2020)	Methylene Blue	Atmospheric Pressure Plasma Jet - Slug Flow Reactor System	Argon, Helium, Oxygen	Sodium Chloride	5,5	2
(Matinzadeh, Shahgoli, Abbasi, Ghoranneviss, & Salem, 2017)	Bromophenol Blue	Atmospheric Pressure Plasma Jet	Argon	-	5,2-2,7	32-43
(Rahimpour, Taghvaei, Zafarnak,	Crystal Violet	Dielectric Barrier Discharge	Oxygen + Argon	Sodium Thiosulphate, Indigotrisulfonate	5,5	-

Referances	Model Dye	Removal Method	Feed Gases	Added Chemical	pH	Conductiviy (µS/cm)
Rahimpour, & Raeissi, 2019)						
(Iervolino, Vaiano, & Palma, 2019)	Methylene Blue	DBD-Cylindrical Configuration	Air, Oxygen	-	6,6	-
(Magureanu, Bradu, Piroi, Mandache, & Parvulescu 2013)	Methylene Blue	Multi-Wire Plate - Pulsed Corona Discharge	Oxygen	Iron (II) Chloride	6	250
(Liu et al., 2016)	Methyl Orange	Atmospheric Pressurized Glow Discharge Plasma	Oxygen	-	-	-
(Pankaj, Wan, Colonna, & Keener, 2017)	Methyl Red	DBD	Oxygen	-	-	-
(Pankaj et al., 2017)	Crystal Violet	DBD	Oxygen	-	-	-
(Reddy & Subrahmanyam, 2012)	Crystal Violet	DBD-Cylindrical Configuration	Oxygen	-	-	-

Source: Prepared by the authors.

Table 5. Comparison of some studies on plasma systems for organic dyes (flow rate, dye concentration, degradation rate, degradation efficiency, energy yield).

Referances	Flow Rate	Dye Concentration (mg/L)	Decomposition Rate (min)	Degradation Efficiency (%)	Energy Efficiency (g/kwh)
(Jiang et al., 2012)	1000 ml/min	60	20	92	11,68
(Chen et al., 2018)	1333,4 ml/min	40	1-90	94	0,0627-5,64
(Chen et al., 2017)	1333,4 ml/min	40	5	97	1,164
(Rashid et al., 2020)	1000	4 4 4	20 60 60	93 86 73	0,863 0,303 0,239
(Chandana et al., 2015)	300 sccm	100	40	Argon 20ppm at MB = 100 Nitrogen 50ppm at MB= 72 Air 50ppm at MB = 88	0,4
(Abdel-Fattah, 2019)	7500 sccm	35	40	98.52	-
(Yamada et al., 2020)	0,2 ml/min	15	3,2	Argon = 89,3 Helium = 75,8 Oxygen = 94,5	Argon = 0,0241 Helium = 0,0226
(Matinzadeh et al., 2017)	5000 ml/min	10	10	-	-
(Rahimpour et al., 2019)	900 ml/min	4,08	1	100	12,24
(Iervolino et al., 2019)	-	10	5	Oxygen = 96 Air = 76	0,79

Referances	Flow Rate	Dye Concentration (mg/L)	Decomposition Rate (min)	Degradation Efficiency (%)	Energy Efficiency (g/kwh)
(Magureanu et al., 2013)	1000 sccm	50	10	98	1,5
(Liu et al., 2016)	-	10	15	93	-
(Pankaj et al., 2017)	-	100	5	100	-
(Pankaj et al., 2017)	-	100	5	100	-
(Reddy & Subrahmanyam, 2012)	-	50-100	25	>89	86,3

Source: Prepared by the authors.

In these studies, it is seen that factors such as the preferred removal method, feed gas used, gas flow rate, added chemicals, dye concentration, degradation rate affect the efficiency of the system. Some of the recent studies on the effectiveness of plasma systems in different configurations in water treatment are given below. Kazemi et al. (2021) investigated the chemical reagent species involved in the degradation of Crystal Violet (CV), a model organic compound, with a new Multiple Array Dielectric Barrier Discharge (MADBD) plasma gas diffuser. Experimental results show that for the best operating conditions, the decolorization rate is 95%, the energy efficiency is 7.8 g/kWh and the CV decolorization rate constant is 2.83 1/min. Investigation of the effect of the relevant reactive species reveals that ozone (O_3) is the most important reactive species causing CV decolorization in MADBD plasma, while short-lived species and reactive nitrogen species have a minor effect on CV decolorization. In their study, Zhou et al. (2021) develop a novel Microplasma Bubble (MPB) Reactor to generate plasma discharges in small formation bubbles as an effective way to provide reactive species for the degradation of target organic pollutants. The combination of plasma with bubbles in the form of underwater microplasma bubbles (MPBs) is observed to enhance the degradation of dyes with high energy efficiency of up to 20 g/kWh. Complete degradation of Alizarin Yellow, Orange-II and Methylene Blue is achieved in 10, 5 and 3 minutes of treatment, respectively. Taghvaei et al. (2019) investigated the chemical reactive species responsible for the degradation of Crystal Violet by Atmospheric Pressure Radiofrequency (RF) Plasma Jet. Different molecular contributions to the argon feed gas (H_2 O, air and O_2) are compared to evaluate the effect of different species produced by the plasma. Argon shows the highest decolourisation effect compared to the other conditions and Ar+0.27% H_2 O leads to the lowest decolourisation effect of Crystal Violet. The results suggest a dominant role of near-surface reactions of short-lived radicals such as OH for the inhomogeneous decolourisation of Crystal Violet. García et al. (2017) prepared a synthetic solution containing Methylene Blue and investigated its degradability by Microwave Plasma Jet. It is found that even with equal total flow and total input power, multiple applicators are slightly more efficient than a single applicator. For equal or lower sample volumes of 50 ml, energy yield values at 50% of dye

conversion are between 0.033 and 0.296 g/kWh, among the best values reported in the literature. The solution becomes colourless after 30 minutes of exposure to the plasma. The plasma reactor was able to produce hydrogen peroxide species in pure water, the concentration of which increased with the plasma argon flow rate. For the most favourable case investigated, the energy yield formation was 2.7mg/(kW-h). Sun et al. (2012) used Pulsed High Voltage Discharge Plasma in their study. Methyl Orange (MO) dye wastewater is selected as the target object. The colour removal rate increases with increasing treatment time. When the initial conductivity is 1700 μS/cm, the colour removal rate is the highest. The degradation efficiency of MO is affected by the gas in the reactor and the degradation efficiency is obtained in the order of oxygen > air > argon > nitrogen and the energy yield obtained in this research is 0.45 g/kWh.

4 CONCLUSIONS

Plasma systems are seen as the technology of the future and an alternative system for the future in line with the studies carried out.

Attri et al. (2016) studied with two needle type direct and indirect atmospheric pressure plasma jets and compared the removal efficiency of three different model dyes (Methylene Blue (MB), Methyl Orange (MO), Congo Red (CR)). Argon feed gas is used in the studies. In 20 and 30 min process times, direct and indirect process results are evaluated separately for three different dyes. In 20 min direct process, MB 95%, MO 97%, KK 86%, in indirect process MB 71%, MO 81%, CR 76% removal efficiency is obtained, in 30 min direct process MB 97%, MO 99%, CR 90%, in indirect process MB 87%, MO 92%, CR 84% removal efficiency is obtained. As a result of the 20 min process, MB 2.8, MO 2.9, CR 2.6, MB 4.3, MO 4.9, CR 4.5 g/kWh are obtained in the direct process, while MB1.99, MO 1.98, CR 1.8, MB 3.4, MO 3.6, CR 3.3 g/kWh are obtained in the indirect process in the 30 min process time. Thus, it can be said that the removal efficiency is higher in the direct process than in the indirect process. In terms of energy efficiency, it is seen that the indirect process is more efficient.

Dojčinović et al. (2011) investigated whether Dielectric Barrier Discharges (DBD) combined with advanced oxidation processes (AOPs) can be used for the decolourisation of reactive textile dyes, namely Reactive Black 5, Reactive Blue 52, Reactive Yellow 125 and Reactive Green 1 5. An effective result of 97% reactive black 5 decolourisation is obtained with the addition of H_2O_2. In conclusion, the results obtained at laboratory level show that the DBD plasma reactor can be effectively used for the decolourisation of textile wastewater.

As a result of the findings obtained, the effect of plasma systems in treatment applications sheds light for the future in the field of Environmental Engineering. In line with the studies carried out, plasma systems contribute to green chemistry applications in terms of allowing the addition of small amounts of chemicals.

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