

Review on pollution reduction from textile industry wastewater by non-thermal plasma technology and its impact on the environment and human

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Abstract: Rapid industrialization and population growth intensify the global demand for clean water, particularly challenging in industries like textiles facing stringent water use permits. This review addresses the textile industry's significant water crisis, emphasizing the role of textile wastewater as a major environmental pollutant, laden with persistent chemicals and heavy metals. Conventional treatment methods, such as adsorption and coagulation, prove inefficient and environmentally hazardous. The review highlights the potential of nanomaterials for contamination removal. As a solution, non-thermal plasma (NTP) technology emerges, demonstrating effectiveness in decontamination. Case studies illustrate NTP's success in achieving over 80% removal rates within short timeframes. Despite NTP's promise, challenges, including energy consumption, must be addressed. Thorough pollutant characterization using various analytical methods, such as ultraviolet–visible (UV–vis) spectroscopy and chromatography, is crucial. The review concludes by emphasizing the need for further research to optimize NTP, making it a cost-effective and sustainable solution for textile wastewater treatment, safeguarding the environment and public health.

Keywords: technology, textile, non-thermal plasma (NTP), wastewater, water, contaminant, pollution

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1 Introduction

Due to the growing population, industry, and agriculture globally, the demand for clean water has surged (Cardoso et al., 2021; Mouele et al., 2021; Palma et al., 2022; Takeuchi et al., 2021). However, the availability of clean water for human consumption is now less than 1%, presenting a critical challenge. Drinking water scarcity is becoming one of the most serious crises worldwide, exacerbated by increasing industrialization and the emergence of pollutants such

as pharmaceuticals, personal care products, and endocrine-disrupting compounds (Cardoso et al., 2021; Mouele et al., 2021; Palma et al., 2022).

Textile industry wastewater (TIWW) stands out as one of the major contributors to water and soil pollution (Kishor et al., 2022). Textile industries consume vast amounts of potable water and use various synthetic chemicals throughout the textile production process (Kang et al., 2020; Ağtaş et al., 2020; Khalish et al., 2022). Liquid waste from the textile industry exhibits common characteristics such as color, alkalinity, high total suspended solids (TSS), biological oxygen demand (BOD), elevated temperature, and certain dyes containing chromium elements. This waste arises from diverse activities,

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including stentering, dyeing, printing, steaming, washing, and finishing (Khalish et al., 2022).

The primary objective of this review is to explore various techniques and technologies for wastewater treatment, emphasizing the effectiveness of non-thermal plasma (NTP) technology as an advanced solution.

2 The amount and impact of textile wastewater on human health and environment

The overview of the different stages of wastewater generation in the textile industry, its toxicity to human health and the environment, and various treatment approaches are summarized in Figure 1 (Kishor et al., 2021; Rame et al., 2021). Many conventional water treatment methods do not completely reduce or remove recalcitrant contaminants, indicating the need for alternatives (KLHK, 2018). Moreover, ongoing climate change will likely increase the demand for water quality management strategies (Kurian, 2021).

Some industries face challenges in fulfilling water needs for production due to restrictive permits for water utilization from local government sources (KLHK, 2018; Zille, 2020). Meanwhile, there is a shortage of new water sources due to reduced discharge (KLHK, 2018).

One of the significant concerns in current studies is wastewater treatment, particularly in the textile industry, as this water impacts human health and the environment, posing carcinogenic, mutagenic, genotoxic, cytotoxic, and allergic threats to living organisms (Kishor et al., 2021; Reema et al., 2022). Different production steps in the textile industry, wastewater generation, toxicity, and various treatment approaches are depicted in Figure 1 (Kishor et al., 2021). TIWW, known for its unpleasant odors, causes river discoloration. In major textile-producing countries, wastewater is often discharged into rivers, which ultimately flow into the sea. For example, in October 2020, numerous dead fish were found along

the Elo River (Khalish et al., 2022; Reema et al., 2022; Bharagava et al., 2018).

TIWW contains a variety of persistent coloring pollutants (dyes), formaldehyde, phthalates, phenols, surfactants, perfluorooctanoic (PFOA), pentachlorophenol, and different heavy metals such as lead (Pb), cadmium (Cd), arsenic (As), chromium (Cr), Zinc (Zn), and nickel (Ni) (Kishor et al., 2021). Heavy metals are employed in the production of color pigments in textile dyes (Rame et al., 2021; Rosa et al., 2020; Senthil Kumar et al., 2023). These pollutants are transported over long distances with wastewater, persisting in the environment for extended periods and posing severe health hazards to living organisms, while also decreasing soil fertility and photosynthetic activity of aquatic plants, leading to toxic conditions for aquatic fauna and flora (Kang et al., 2020).

Some research implies that dyes used in the textile industry, such as Methylene blue, are highly dangerous, exhibiting carcinogenic and mutagenic effects with strong chemical structures matching emerging contaminants (ECs) (Joshi et al., 2022; More et al., 2020).

Textile manufacturing involves different complex processes. The various stages of textile manufacturing processes are as follows (Kishor et al., 2021; Fazal et al., 2020):

(1) Sizing

It is the initial step in textile production from man-made or natural fibers like polyester, silk, jute, cotton, and wool. Special substances such as carboxymethyl cellulose (CMC), polyvinyl alcohol (PVA), polycerate, and polycyclic acids are added to provide high potency to fibers.

(2) Desizing

The second step utilizes enzymes and various auxiliary chemicals to remove unwanted chemicals and sizing materials, enhancing the absorbency of fibers. Bacterial enzymes and mineral acids are more prevalent than traditional methods in desizing applications.

(3) Scouring

A cleaning process used to remove impurities from fibers, alkali agents like glycerol, ethers, sodium hydroxide, detergent, or soap are used for removal and washing of impurities such as fats, waxes, oils, surfactants, and non-cellulosic materials.

(4) Bleaching

A chemical process used to remove unwanted coloring materials from fibers. Currently, H₂O₂ and peracetic acid are used as bleaching agents to enhance the whiteness of fibers.

(5) Mercerization

Cold or hot caustic soda (NaOH) is extensively used in this process to improve the physical and chemical properties of fibers, including luster, strength, dye affinity, and fiber appearance.

(6) Dyeing and Printing

A variety of auxiliary chemicals are used at this stage to improve the attachment of dye molecules with fibers. Dyeing is a major process in textile production for adding colors to fabrics, with various dyes extensively used worldwide. Phthalates, dyes, metals, solvents, formaldehyde, and urea are commonly used in the printing stage.

(7) Finishing

The final stage involves the use of different types of protection and maintenance chemicals, such as biocides, and synthetic organic or inorganic chemicals, to improve and maintain specific fiber properties, including stain proofing, softening, waterproofing, flame retardance, and protection from microbial activities as well as ultraviolet (UV) damage.

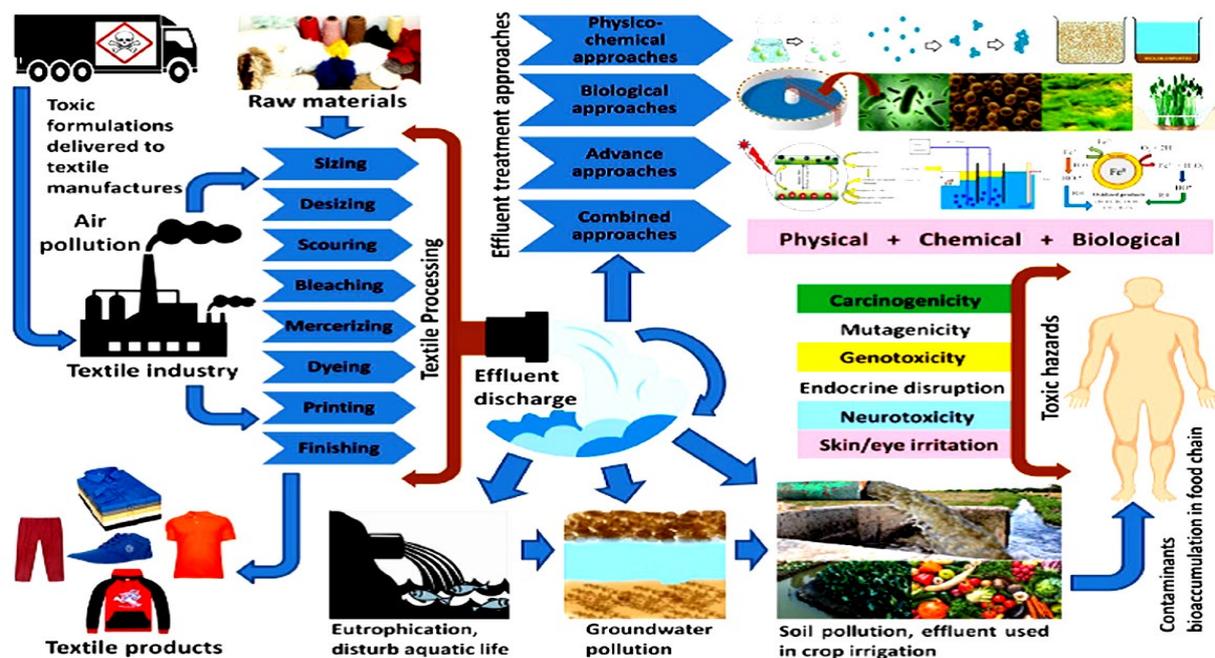


Figure 1 Processing in the textile industry, wastewater generation, its toxicity, and various treatment approaches (Kishor et al., 2021)

Dyeing and washing in textile industries are the major sources of wastewater generation. A typical textile industry uses around 1.6 million liters of groundwater to produce 8000 kg of textile fabric per day (Garg et al., 2020). Almost 30%-40% of water is consumed in the dyeing process, 60%-70% in the washing stages, and approximately 10%-50% of unused dyes are released into resources along with the generated wastewater (Garg et al., 2020). Dyes

such as methyl orange, congo red, methylene blue, azure B, reactive dye, direct red, remazol red, scarlet, malachite green, acid orange, and remazol brilliant blue R are extensively used in textile industries (Cai et al., 2020). Textile dyes are classified based on molecular structures, charges, and their potential health effects (Pavithra et al., 2019).

Some significant hazardous water contaminants for human health, such as endocrine inhibitors and

chlorine, are treated by-products (Jakob et al., 2023). Other chemical elements known to be seriously dangerous include phenols, which have highly toxic compounds causing irritation when in direct contact with the skin. Phenols in water can affect tissues in fish and animals directly (Sugiharto, 2014).

Despite the need for improvement in wastewater treatment plant effluent water quality and the efficient installation of reuse units to meet reuse requirements (KLHK, 2018), new treatment methods should be sought with advantages such as safety, convenience, and lack of residual toxicity (Al-Rawaf et al., 2018).

3 Current status of wastewater treatment in textile industry

There are numerous techniques and technologies for wastewater treatment discussed in this review as effective methods. For instance, conventional water treatment technologies encompass coagulation, sedimentation, filtration, ozonation, and disinfection using chlorine (Takeuchi et al., 2021; Stefan 2018).

3.1 Physico-chemical methods

Physico-chemical methods, such as adsorption and coagulation, prove effective for color removal from TIWW (Kang et al., 2020). The adsorption method transforms pollutants from one phase to another, while coagulation/flocculation methods selectively decolorize dyes like sulfur and dispersive dyes. However, they are not useful for the decolorization of acid, reactive, direct, and vat dyes (Kang et al., 2020). These methods are expensive, time-consuming, less applicable, and produce large amounts of very toxic sludge as a secondary pollutant, posing significant risks to the environment, humans, and animal health (Kang et al., 2020; Pavithra et al., 2019).

3.2 Wetlands construction

Wetlands construction emerges as a robust method against wastewater pollution, preventing issues for the biotic environment. Constructed wetlands offer an alternative to planned and controlled water treatment, utilizing natural processes involving vegetation, media, and microorganisms

(Nurmitha, 2017). This method is known for its flexibility in the operation system, ease of construction, flexibility in selecting placement locations, and cost-effectiveness (Nurmitha, 2017).

3.3 A reactor with mixed aquatic plants, single aquatic plants, and single aquatic plants

A study conducted in Pringsurat subdistrict, Magelang Regency, Central Java province, aimed to evaluate the performance of textile industry units. Water quality analysis was based on parameters such as total phenol, ammonia, and sulfide, following Central Java provincial regulation No. 5, 2012, on Wastewater Quality Standards (Khalish et al., 2022). Three different treatments with three types of reactors were employed: a reactor with mixed aquatic plants, single aquatic plants, and single aquatic plants (Khalish et al., 2022). Laboratory tests on 7 parameters of textile waste, including high TSS, BOD, chemical oxygen demand (COD), phenol, chrome, sulfide, and ammonia, indicated that five of them exceeded the mentioned standards—BOD, COD, phenol, ammonia, and TSS. The high values of COD, BOD, and TSS reveal that TIWW contains a significant amount of chemically oxidizable substances and high organic pollution from the textile industry process (Khalish et al., 2022). The dyeing process also generates more solid waste (Khalish et al., 2022). The research results showed that *Typha angustifolia* reactors exhibited significantly higher processing efficiency for the dominant parameters, resulting in reductions of 88.98%, 80.82%, 89.51%, 89.81%, 79.95%, and 96.71%, respectively (Khalish et al., 2022). These reactors successfully processed 5 out of the 6 tested parameters to meet the specified quality standards, including BOD, COD, TSS, total phenol, and sulfide. In contrast, the *Iris pseudacorus* reactor demonstrated less effective processing, meeting quality standards for only 2 out of 6 parameters (Table 1) (Khalish et al., 2022). This disparity may be attributed to suboptimal plant adaptation and the plant's age not reaching an optimal level for efficient phytoremediation (Khalish et al., 2022). Furthermore, the test results indicated that

Typha angustifolia plants performed better when used alone compared to their combination with *Iris pseudacorus* (Khalish et al., 2022). This discrepancy may arise from the suboptimal waste processing

capabilities of *Iris* plants or an incompatible interaction between the two plant types, potentially leading to competition in pollutant absorption from wastewater (Khalish et al., 2022).

Table 1 Wastewater quality test results with constructed wetland (Khalish et al., 2022)

Parameter	Unit	Iris pseudacorus		Typha Agustifolia		Combination	
		Concentration	Efficiency (%)	Concentration	Efficiency (%)	Concentration	Efficiency (%)
BOD	mg L ⁻¹	91.3	69.3	32.5	88.98	70.0	76.27
COD	mg L ⁻¹	172.5	64.24	92.5	80.82	131.6	72.72
TSS	mg L ⁻¹	16	88.8	15	89.51	14	90.2
Total Phenol	mg L ⁻¹	<0.0215	89.81	<0.0215	89.81	0.0383	81.84
Total Chromium	mg L ⁻¹	<0.0095	-	<0.0095	-	<0.0095	-
Ammonia (NH ₃ as N)	mg L ⁻¹	76.7519	60.81	39.2500	79.95	53.3927	72.73
Sulphide (as S)	mg L ⁻¹	<0.0043	96.71	<0.0043	96.71	<0.0043	96.71

The combination reactor exhibits an intermediate level of efficiency compared to the other two reactors (Khalish et al., 2022). This suggests that the use of a combination of two types of plants does not lead to an increase in the efficiency of pollutant removal, likely due to competition between the two plant types (Khalish et al., 2022).

3.4 Nanomaterials technique

The use of nanomaterials, such as nanoparticles, nanomembranes, and nanotubes, has proven to be quite effective for the detection and removal of various chemical and biological substances, including metals, algae, organic substances, bacteria, viruses, nutrients, and antibiotics. Nanotechnology describes the characterization, fabrication, and manipulation of structures, devices, or materials that have one or more dimensions smaller than 100 nanometers (Cardoso et al., 2021; Baig et al., 2021). Currently, nanomaterials that have proved important for the degradation of organic matter in wastewater are classified as dendritic polymers, metal/metal oxide nanoparticles, zeolites, and carbon-based nanomaterials (Cardoso et al., 2021). The coupling of an intrinsic material, polyether sulfone nanofibers (PES NF), resulted in an efficient adsorption-photocatalytic degradation nanosystem for methylene blue dyes (Homaeigohar et al., 2019).

TiO₂ nanoparticles can be employed to catalyze

the degradation of organic compounds and dyes, frequently encountered in textile wastewater (Senthil Kumar et al., 2023). TiO₂ represents the chemical formula for titanium dioxide, a naturally occurring oxide of titanium (Senthil Kumar et al., 2023). TiO₂ is a white, odorless, and tasteless powder frequently utilized as a pigment in paints, coatings, plastics, and various other materials (Senthil Kumar et al., 2023). This method in wastewater treatment should be done carefully, taking into consideration all sustainability indexes, and particularly ensuring that it does not result in damage to the environment (Adeleye et al., 2016). Nanomaterials will be part of the solution for future water management strategies. Of course, nanomaterials will enhance performance because fewer resources are used for their production, and greener catalytic processes are implemented (Cardoso et al., 2021).

3.5 Enzymes method

Many enzymes are reported to engage in the degradation and detoxification of industrial wastewater pollutants, as well as remediation sites. Enzymes such as lignin peroxidase, azoreductase (a major class of azo dye degrading enzyme), and laccase are also reported to play important roles in the degradation of various pollutants from TIWW. Azoreductase, by catalyzing the cleavage of azo linkage under aerobic and anaerobic conditions, can

degrade and decolorize several azo dyes. The advantages of enzymatic treatment are eco-friendly and have the possibility to influence the complete remediation of pollutants. However, the disadvantages consist of long-period treatment, sensitivity to temperature and pH, inactivity against toxic compounds, and inapplicability at a large scale (Kang et al., 2020; Zhuang et al., 2020).

3.6 Ultrafiltration technique

Ultrafiltration (UF) membrane, due to its low energy consumption, has become an alternative technology in water and wastewater treatment (Sidabutar et al., 2020). However, there is a limitation in the conventional application of this treatment method: its low rejection of soluble compounds, for instance, dyes in textile wastewater. Polysulfone has widely been employed in UF membrane preparation because of its good mechanical strength and good membrane-forming performance (Rameetse et al., 2020). In this study, different concentrations of polymer were investigated for their impact on membrane performance (Rameetse et al., 2020). During the textile wastewater treatment, the profile of permeate flux was observed. For this experiment, the membrane selectivity was tested against naphthol-AS, which was employed as a coupling partner in some azo dyes preparation in the textile industry, and for analyzing a UV-Vis spectrophotometer at a wavelength (λ) of 294nm was used. Finally, the result depicts that the UF membrane that was prepared to blend 20 wt.% polysulfone with 20 wt.% polyethylene glycol and 1 wt.% acetone resulted in better performance compared to 16 and 18 wt.% of polysulfone. A high polysulfone content decreased irreversible (internal) fouling in the membrane structure. However, improving the membrane selectivity needs another optimization (Aryanti et al., 2021).

3.7 Advanced oxidation processes (AOPs) methods

Although chlorine is one of the most used chemical oxidants worldwide for disinfection of drinking and wastewater contaminants, it does not work for many contaminants (Jakob et al., 2023; Bu

et al., 2018). Water treatment processes employing OH are called AOPs (Takeuchi et al., 2021). Currently, many AOPs, including ozonation, photo-Fenton, photocatalytic, sono coagulation, electro-coagulation, and electro-oxidation processes, are widely employed in the degradation and mineralization of different persistent dyes, dissolved solids, and heavy metals from TIWW (Cardoso et al., 2021; Yamada et al., 2020). However, these are expensive, have high chemical demand, use complicated procedures, apply high electrical energy, and generate huge amounts of sludge as a secondary pollutant (Paździor et al., 2019). For instance, conventional ozone processing based on AOPs has excellent disinfection capabilities through the action of the hydroxyl radical (OH) and works much better than chlorine, an oxidizing pollutant present in wastewater and industrial effluent (Cardoso et al., 2021). AOPs, compared to conventional water treatment techniques, have low temperatures, operate easily and stably at atmospheric pressure, greater efficiency and capacity to degrade recalcitrant organic pollutants, and can generate fewer toxic intermediate products during their degradation, but they are not known as an economic method (Mouele et al., 2021; Jakob et al., 2023; Yamada et al., 2020; Domingues et al., 2021).

3.8 Catalytic ozone technology

The other technology that has been employed in industries for wastewater treatment is catalytic ozone technology, but this technology remains energy-consuming and, as a consequence, expensive (Rame et al., 2021). Catalytic ozone methods have proven to be useful for eliminating organic contamination in wastewater, to the extent that it could reach the effluent standard for reuse (Permenkes 32/2017, clean water standard). An experiment was done for the degradation of wastewater by using this technology (Rame et al., 2021). So, in this project, a reactor consisting of four main parts was employed (Rame et al., 2021). The parts were a continuous-mode catalytic ozone reactor, a filtration unit, a solar panel for electricity supply, and a wastewater real-time

monitoring controller to analyze flow and effluent parameters such as pressure, flow level, temperature, etc. (Rame et al., 2021). The capacity of this reactor was 50 per day. Finally, the combination of catalytic ozone and sensor integration in the real-time process can produce good-quality water for the industry. On the other hand, the mixture became a relatively expensive process for the industry because of the energy/electricity requirement from fossil fuel-based electricity (Rame et al., 2021).

The treatment of TIWW is a major challenge, as there is no particular economically feasible treatment method capable of adequately treating TIWW (Yamada et al., 2020). Therefore, there is a need to develop a novel, cost-effective, and eco-friendly technology for the effective treatment of TIWW (Takeuchi et al., 2021; Jakob et al., 2023).

3.9 Plasma treatment

Plasma treatment has been investigated as an effective and useful decontamination method because the goal of plasma water treatment, in comparison with the conventional ozone method, is to move the plasma source adjacent to water. Along with ozone, during the ionization, free-radical species are produced by the plasma discharge (Jakob et al., 2023). This technology is technically a green process with no generation of chemically harmful substances and has more ecological and economic benefits (Reema et al., 2022). When plasma comes in contact with water, OH is generated from water molecules via electron-impact dissociation and reaction with radicals, such as O radicals and metastable atoms, such as Ar metastable atoms (Takeuchi et al., 2021). So, a portion of the gas-phase OH diffuses into water and decomposes organic compounds (Takeuchi et al., 2021).

4 Opportunity, advantages, and challenges of NTP in wastewater treatment

For decades, electrical discharges (EDs) such as corona discharges (CD), dielectric barrier discharges (DBD), atmospheric pressure plasma jet (APPJ), and micro-hollow cathode discharges (MHCD) have been

considered as the major types of ANTPs regardless of their characteristic properties and applications (Mouele et al., 2021; Khanikar et al., 2021; Takeuchi et al., 2021). These discharge processes have emerged in recent years as effective methods for water decontamination (Mouele et al., 2021).

Most importantly, plasma technologies are capable of generating a mixture of highly reactive species and radicals under atmospheric conditions, which is beneficial in organic and other industrial treatment processes (Ogunyinka et al., 2020).

Ozone is one of the most utilized oxidants for water purification following various reaction mechanisms or reaction pathways due to the existence of various reaction sites on organic pollutants (Mouele et al., 2021).

One of the most challenging tasks during wastewater treatment by plasma technologies is the determination of reactive oxygen species (ROS) in aqueous plasma discharge actuators because of their inadequate selectivity and limited lifespan (Mouele et al., 2020b).

Research in the field of plasma's technological applications concerns two main issues (Stryczewska, 2020):

(1) New solutions of plasma reactors (PRs) and electric discharges used in them, in which plasma, with the required plasma-chemical parameters and high time-space effectiveness, will be produced efficiently at atmospheric pressure.

(2) Efficient and controllable plasma reactor power supply systems with a wide range of changes in the properties (voltage shape and frequency, range of regulation), which are an inseparable part of the plasma-chemical installation that determines industrial implementation.

The development of cost-effective wastewater treatment techniques is crucial for sustainable water resources. NTP processes like DBDs are being used to remove stubborn organic substances in water. They generate UV light and various ROS like ozone (O₃), hydrogen peroxide (H₂O₂), and others for effective treatment (Cardoso et al., 2021; Mouele et al., 2021;

Al-Rawaf et al., 2018; Mouele et al., 2020a). Unlike other types of plasma atmospheric non-thermal plasma (ANTP) occurs at a temperature range of 300–50,000 K. ANTP is characterized by its electron temperature (T_e) which is much higher than the initial temperature (T_i) but is in turn approximately equal to gas temperature (T_g) that lies between 300 and 1000 K ($T_e \gg T_i > T_g \approx 300 \cdot 10^3$ K) and a corresponding ionization energy range of 1–10 eV (Cardoso et al., 2021; Huang et al., 2018; Russo et al., 2020). Besides, ANTPs are also featured by their electron density (n_e) estimated to about $10^9 - 10^{15} \text{ m}^{-3}$ ($10^9 \leq n_e \leq 10^{15} \text{ m}^{-3}$) (Mouele et al., 2021).

As an example of a plasma system, the DBD system also generates UV light, O^3 , and H_2O_2 , and their combination in aqueous media allows water and wastewater treatment without additional chemicals (Mouele et al., 2021; Takeuchi et al., 2021; Zahoranová et al., 2018). So far, it is showing positive results which increase non-thermal plasma as an environment-friendly technology (Reema et al., 2022; Al-Rawaf et al., 2018). Electrode geometries may affect these systems' efficiency (Mouele et al., 2021).

Plasma in an aqueous solution generates ROS and RONS which are mainly responsible for the inactivation of pathogens and bacteria (Reema et al., 2022).

Forming new radicals such as OH that may directly or indirectly attack the targeted pollutants. Ozone can also be combined with catalysts and UV light to initiate photocatalytic phenomena (Mouele et al., 2021). In addition, during the treatment of drinking water, the speed of reaction of ozone with various inorganics, ionized and dissociated inorganic components are extremely high (Mouele et al., 2021; Reema et al., 2022).

Plasma technology has many uses in the textile industry. It can make fabrics stronger, prevent static electricity, improve how well they absorb liquids, make dyeing and printing more eco-friendly, turn regular fabrics into flame-resistant ones, and help

things stick to the fabric better (Reema et al., 2022).

Using plasma technology in textiles could change how things are usually done in the industry. One big problem in making textiles is that dye often gets wasted and harms the environment. Plasma technology can help by cleaning fibers and making it easier for dye to get into them. This makes colors brighter and last longer even after washing. It also reduces the amount of dye and water needed. Plus, it makes dyeing more consistent, saves money, and helps the environment by reusing some of the waste (Reema et al., 2022).

Combine treatment approaches using physical and biological methods can be used for better degradation and mineralization of TIWW in comparison with other methods in a single use (Yamada et al., 2020; Sun et al., 2020). For example, Sun et al. assessed a combined system of plasma oxidation and microbial fuel cell (MFC) and found it effective to remove and mineralize 97.7% of methylene blue dye with the generation of electrical energy up to 519 mWm^{-2} (Sun et al., 2020).

5 Reactor design guided by modeling, and construction

NTP, commonly known as “cold”, has been used for approximately two hundred years. At the beginning of the 19th century, PRs with barrier discharge were introduced as a treatment technology for drinking water, first in Nice (1907) and then in Saint Petersburg (1908) (Reema et al., 2022; Stryczewska., 2020). Nowadays, NTP treatment is widely used in environmental processes of air, water, and soil decontamination, as well as in biology and medicine. For example, the removal of hazardous organic pollutants from wastewater involves pulsed corona discharges and DBDs. Different kinds of reactive radicals such as O_3 , H_2O_2 , OH, etc., are produced when plasma technology is used in wastewater treatment (Yamada et al., 2020). OH radicals are known as strong oxidizing active species for dye decolorization, such as Methylene Blue (Yamada et al., 2020).

In laboratories, plasma can be produced by various techniques and mainly by supplying electrical energy to gas (Reema et al., 2022). When adequate voltage is applied between two electrodes, it can cause a breakdown of the gas, and plasma is formed (Reema et al., 2022). As long as the applied voltage exceeds the breakdown voltage, which varies for different gases, plasma will continue to flow (Reema et al., 2022).

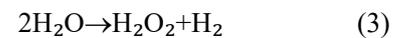
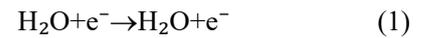
Plasma reactor's macroscopic characteristics are strongly non-linear, and their work is commonly carried out at high AC, DC, or pulsed voltage, often with increased or high frequency (Reema et al., 2022; Stryczewska, 2020). In industrial use, PRs need a high-power device, with mains requiring additional devices such as reactive power compensation systems or filters, reducing distortion of the mains current. To choose and design a plasma supply system for a given plasma process, one needs to specify the requirements and parameters of the plasma receiver, the most important of which are (Stryczewska, 2020):

- Supply voltage—constant, sine, impulse;
- The presence of an additional ignition system of the discharge or its absence;
- NTP reactor power and attainable power of the PSS;
- The ability to adjust the current value and maintain its continuity in the entire area of the plasma reactor operation;
- The ability of the power source to work in automatic control and regulation systems and adjustment of parameters to various process gases and their mixtures;
- Correct cooperation with the power supply network;
- High efficiency;
- Simplicity and safety of use;
- Low capital and operating costs.

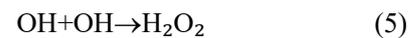
So, these numerous requirements make NTP reactors' supply systems complex (Stryczewska, 2020).

The following reaction formulas represent simple pathways for the generation of active species via

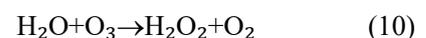
electrical discharge plasma in water (Lukes et al., 2011).



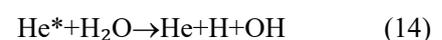
At a lower electron energy, reaction (1) primarily occurs, whereas reaction (2) is dominant at a higher electron energy (Lukes et al., 2011). Although these radicals easily return to the stable ground states, it is presumed that they may combine to form other molecules such as hydrogen (H_2) and hydrogen peroxide (H_2O_2) as represented in reaction (3) (Lukes et al., 2011). The reaction formulas (4)– (6) denote the formation of molecules from active states species via the dissociation of water molecules (Lukes et al., 2011). In particular, reaction (5) is important because short lifetime OH radicals are converted to long lifetime H_2O_2 molecules (Lukes et al., 2011).



Oxygen, helium, and argon are known as common feed gases in wastewater treatment by NTP technology. So, the reactions via oxygen plasma are as follows (7)- (10) (Yamada et al., 2020).



And, the electron impact reactions that were caused by argon and helium gases and OH radical generation at the plasma discharge are shown in the reaction formulas (11)– (14) (Yamada et al., 2020).



Takamatsu et al. investigated OH radical production in an aqueous solution irradiated by an atmospheric-pressure non-equilibrium plasma jet (Takamatsu et al., 2014). They determined that the introduction of argon gas for the plasma jet source

caused a higher amount of OH radical production compared to those of oxygen and helium gases, respectively. Therefore, it is possible to assume that the abundance of OH radical generation might promote the decolorization of the Methylene Blue dye (Takamatsu et al., 2014).

NTP reactors with dielectric barrier discharge (DBD) and APPJ require supply systems (Stryczewska, 2020). Since O_3 and H_2O_2 are produced in DBD systems, the addition of semiconductor photocatalysis may significantly improve water treatment (Mouele et al., 2021). These types of discharge produce simple, and the potential area of application is large, especially in environmental protection processes. In improving biotechnologies, these kinds of reactors can be powered from similar sources using nonlinear transformer magnetic and power electronics systems (Mouele et al., 2021).

DBD Systems: The combination of DBD systems is an effective route to enhance the concentration of reactive species and, hence, the efficiency of the DBD configuration for efficient degradation of water contaminants (Ren et al., 2021). In NTP technologies induced by DBD, as with any other AOPs, OH radicals react with organics in four different ways,

namely, hydrogen abstraction, radical addition, electron transfer, and radical combination (Mouele et al., 2021).

The creation of plasma with the DBD method, due to its economic benefits, simple arrangement, and adaptability for the electrode shape and dielectric material used, is gaining significance (Reema et al., 2022).

The DBD method is gaining importance in plasma technology because it's cost-effective and versatile. It's considered valuable because it's easy to set up and can adapt to different electrode shapes and dielectric materials (Reema et al., 2022). In DBD, plasma is created by applying high voltage between two metal electrodes - a high-voltage terminal and a ground electrode, as shown in Figure 2. These electrodes are often surrounded by materials like glass, quartz, polymer, mica, or even artistic materials (Reema et al., 2022). The gap between them can vary from very tiny (0.1 mm) to a few centimetres, and in the resulting argon plasma, you find a high electron density of about 10^{15} cm^{-3} (Reema et al., 2022). Typical operating conditions involve gas pressures between 10^4 and 10^6 Pa, frequency bands from 10 to 50 MHz, and voltage levels ranging from 1 to 100 kV_{rms} for alternating current (Feizollahi et al., 2021).

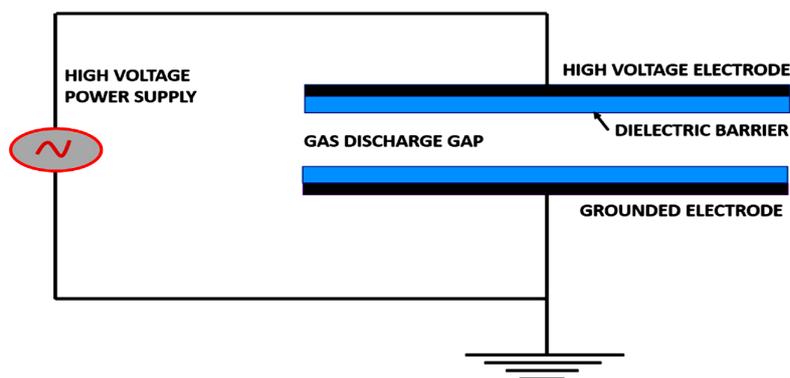


Figure 2 Schematic of dielectric barrier discharge—DBD (Reema et al., 2022)

Air Discharge Plasma Jet: Air discharge plasma jet has been utilized for the treatment of textile wastewater (Mouele et al., 2020a). The common feed gases used in DBD advanced technologies include dry air, helium, nitrogen, oxygen (O_2), and argon (Ar) (Reema et al., 2022; Wardenier et al., 2019; Coutinho et al., 2018). Some researches show that

excellent decomposition of model toxins could be achieved with plasma followed by argon and air plasma. This classification is likely associated with the nature and different amounts of chemical species produced in each system at ambient conditions (Crema et al., 2020).

The most common design uses coaxial electrodes

with gas flowing between them. In this setup, a discharge occurs between a grounded electrode and a live electrode, as shown in Figure 3. When power is applied, a discharge ignites and operates on a feed gas passing between the outer grounded electrode and the live electrode, resulting in the creation of a discharge beam (Reema et al., 2022). In this process, free electrons are accelerated and collide with gas molecules, producing reactive species. A high gas flow rate is maintained between the electrodes, and the resulting plasma flows into the open air with the

gas. Plasma jets offer a consistent, stable, and uniform release of plasma at atmospheric pressure (Reema et al., 2022). Typical plasma parameters like electron density and temperature for CAP (cold atmospheric plasma) devices typically range from 10^{13} to 10^{15} cm^{-3} and 1–10 eV, respectively (Reema et al., 2022; Wang et al., 2020). Due to its simplicity and the production of cold plasma, it finds wide applications in the biomedical field (Reema et al., 2022).

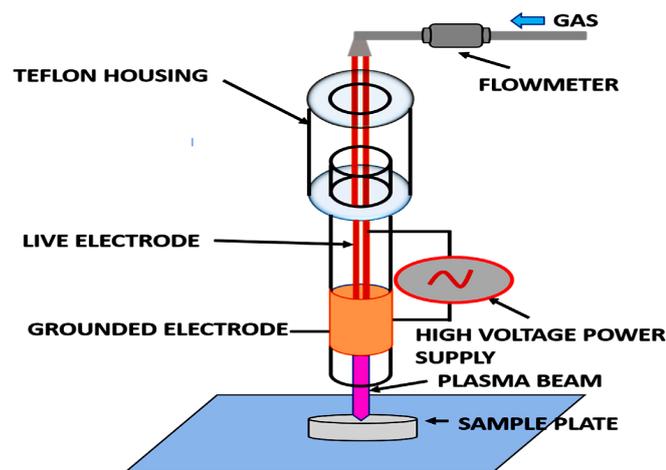


Figure 3 Laboratory set up of cold plasma (CAP) at atmospheric pressure (Reema et al., 2022)

The mechanism of bacteria inactivation in a plasma jet can occur through two known factors: physical and chemical factors (Al-Rawaf et al., 2018). The physical factors include heat, UV irradiation, and discharge particles, while the chemical factors encompass the active species (Al-Rawaf et al., 2018).

In the study that employed the DBD plasma jet system under atmospheric conditions against ECs in the textile industry, such as Rhodamine B and Methylene Blue used as dyes, an AC power supply of 0-6.5, 25 kHz was utilized to generate the plasma. For this process, a glass plasma reactor with an inner and outer diameter of 4 mm and 6 mm, respectively, was employed (Joshi et al., 2022). A 1.5 mm diameter copper rod served as a high-voltage electrode, and a 1 mm diameter copper wire was wrapped around a glass tube as a ground electrode. Argon and helium gases were used one by one for the generation of plasma. The schematic of this system is shown in Figure 4 (Joshi et al., 2022).

So, in this project, they considered each section that could affect this experiment, such as pH, conductivity, and the sample's distance from the plasma (Joshi et al., 2022). Eventually, they achieved removal rates exceeding 80% within the first 5 minutes and completed removal within 30 minutes (Joshi et al., 2022). They concluded that both argon and helium gases yielded similar results but recommended argon due to cost-efficiency (Joshi et al., 2022).

Figure 5 shows the experimental setup for the plasma microbubble, consisting of the plasma bubble reactor, the high-voltage power supply system, and the air supply system. The focus of this study was on the degradation efficiency of methylene blue (MB), which is commonly used as a general dye and for biological staining. It is known as a water pollutant found in most industrial wastewater (Fernandez-Perez et al., 2020). In this system, the plasma bubble reactor has a fully sealed air intake enclosure that bubbles

into a solution container. Plasma is generated on the porous surface DBD plasma source in the region between the plasma source and membrane (Fernandez-Perez et al., 2020). The water comes into contact with the plasma when the air inflows into the bottom enclosure, forcing reactive species generated

in the plasma through the membrane and generating microbubbles. The size and formation of the generated microbubbles depend on membrane characteristics, such as pore size or the wettability of the material (Fernandez-Perez et al., 2020).

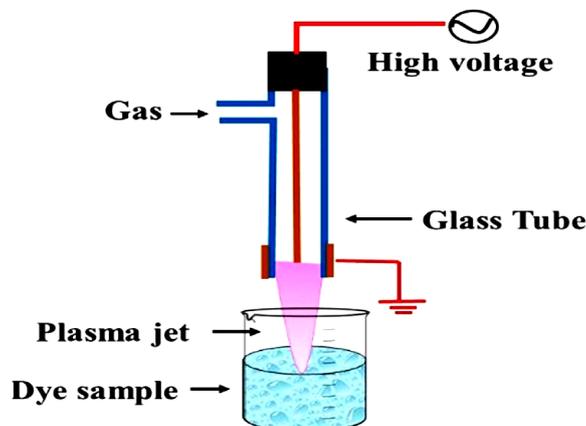


Figure 4 Schematic DBD plasma jet treatment system (Joshi et al., 2022).

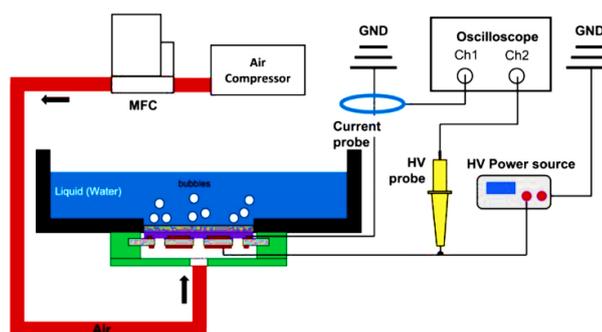


Figure 5 The experimental setup for the plasma microbubble system by DBD plasma (Fernandez-Perez et al., 2020).

In this study, the sinusoidal signal is step-wise amplified through the audio amplifier and the high-voltage transformer, and the highest amplitude of the voltage signal output goes up to 4.5 kV. The frequency of the signal stays steady at 6 kHz. The air supply system consists of a mass flow controller providing an air flow rate ranging from 0 to 12 standard liter per minute (slpm) (Jakob et al., 2023). Finally, in this project, different processing parameters (treatment time, solution volume, initial concentration, electrode-filter distance, and gas flow rate) were assessed for this plasma configuration (Reema et al., 2022; Jakob et al., 2023; Takeuchi et al., 2018). The result depicts that a degradation efficiency of over 90% was achieved after 5 minutes of pollutant water treatment (Reema et al., 2022; Jakob et al., 2023). Varying initial MB concentration and distance between the membrane

and the plasma source did not affect exponential degradation. However, different solution volumes decreased in degradation efficiency with increasing volume, suggesting that the reaction rate increased with the increase of MB in the system (Jakob et al., 2023). An increase in air flow rate can increase degradation efficiency, and because of that, the added reactive gases increase the reaction rate of degradation. This project illustrates how the operating parameters of the plasma bubble reactor can be optimized to improve the MB degradation performance. Overall, the various parameters of the reaction rate have been linked back to their changes in the quantity of reactant in the system (Jakob et al., 2023).

In another study, a group developed an atmospheric pressure pulsed discharge plasma process in a slug flow system (Yamada et al., 2020).

In this study, they used an apparatus that consisted of a glass column (2 m×1.8 mm), high-performance liquid chromatography, a gas flow meter, and a power supply (Yamada et al., 2020). In this experiment, Methylene blue was selected as the organic dye compound because it has an intense color and is used

in industries such as textiles. Oxygen, helium, and argon were employed as feed gases for discharge plasma generation (Yamada et al., 2020). Figure 6 shows a schematic of the experimental apparatus (Yamada et al., 2020).

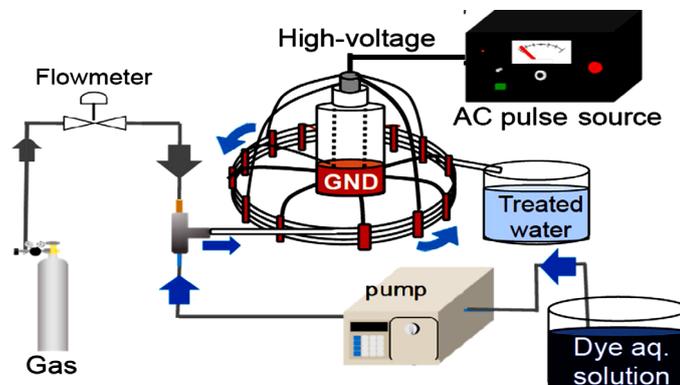


Figure 6 A schematic of the experimental apparatus (Yamada et al., 2020)

During this experiment, the liquid and the gas flowed simultaneously from different directions and were mixed. The gas-liquid slug flow, with the same interval of bubbles, was formed by tuning their flow rates in a capillary glass column reactor. Furthermore, the residence time of the bubbles was approximately 200 seconds. The results show that more than 90% of

the decolorization rate was achieved for one-time plasma treatment with various gas species, with the Methylene Blue dye decolorization rate order being oxygen > argon > helium (Yamada et al., 2020).

The experimental setup presented in Figure 7 was applied for treating acetic acid solution (Takeuchi et al., 2021).

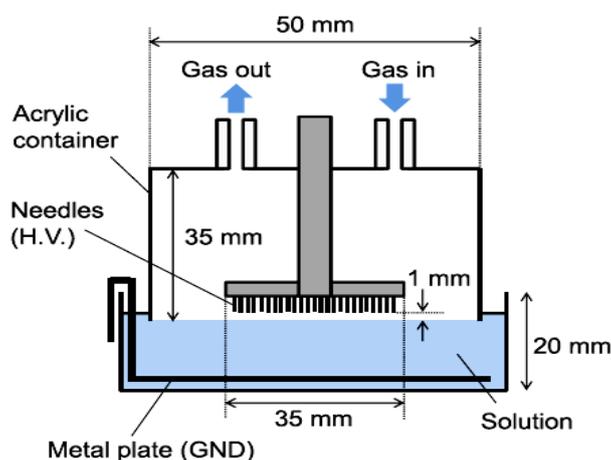


Figure 7 Schematic of the reactor used for plasma generation over a solution (Takeuchi et al., 2021)

A metal disk with 84 attached needles is secured above the solution (20 ml). There exists a 1 mm gap between the tips of the needles and the surface of the solution (Takeuchi et al., 2021). The initial TOC concentration was 10.7 mg TOC L⁻¹. A pulsed voltage is applied to the high-voltage (H.V.) electrode through a 109 Ω resistor with Ar gas to generate filamentary plasma channels over the solution surface.

It has been verified that acetic acid was decomposed into CO₂ gas via the generation of formic acid and oxalic acid as byproducts in the solution (Takeuchi et al., 2021).

As a result, a diaphragm discharge plasma, capable of achieving a higher production rate of H₂O₂ at a greater energy efficiency within gas bubbles, was integrated with an ozonizer (Takeuchi et al., 2021).

Figure 8 illustrates a diaphragm discharge plasma reactor utilized in conjunction with an O₃ supply (Takeuchi et al., 2021).

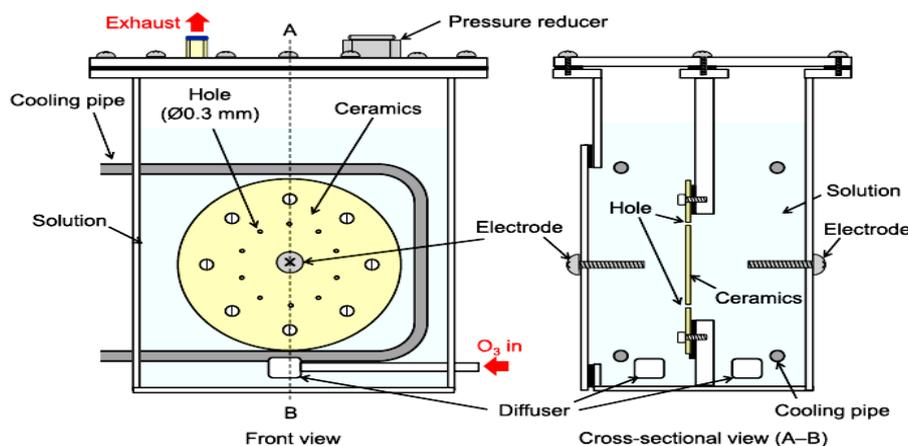


Figure 8 Schematic of the combination process using diaphragm discharge plasma with ten holes and an ozonizer (Takeuchi et al., 2021)

This reactor comprises two solution containers separated by a ceramic plate with ten small holes (0.3 mm in diameter), and an electrode in each container. An AC rectangular voltage is applied between the electrodes to generate diaphragm discharge plasma in the holes (Takeuchi et al., 2021).

6 Test, verification and validation

These metabolites necessitate thorough characterization and identification through a variety of analytical methods to gain insights into their properties, all in the interest of safeguarding the environment and public health. UV-vis spectroscopy is the primary tool for assessing textile dye decolorization (Mouele et al., 2021; Yamada et al., 2020; Huang et al., 2022). Prominent absorbance peaks in the visible light spectrum disappear, and new peaks emerge as evidence of successful dye removal (Kishor et al., 2021). The American Dye Manufacturers Institute (ADMI) tristimulus filter method is used to assess decolorization (Kishor et al., 2021). Fourier transform infrared (FTIR) analysis identifies functional groups in original compounds and metabolites (Mouele et al., 2021; Takeuchi et al., 2021; Kishor et al., 2021). High-performance liquid chromatography (HPLC) is used to detect, identify, and quantify organic compounds (Kishor et al., 2021). GC-MS and LC-MS/MS are used for characterizing low molecular weight organic compounds and their

metabolites (Kishor et al., 2021). Nuclear magnetic resonance (NMR) confirms the presence and proton positions in organic compounds and metabolites (Kishor et al., 2021).

An advanced laser induced fluorescence (LIF) system assembled with a self-developed fluorescence telescope, an optimized and synchronized tunable pulsed laser with high precision, followed by a surface discharge generator and intensified charge coupled device (iCCD) camera, and an oscilloscope has also been reported effective for the detection and measurement of RNS (~ ppb level) in DBD systems (Gao et al., 2017).

For the quantification of the dyes decolorization rate by plasma discharge can use the following Equation 15 (Yamada et al., 2020):

$$\text{Decolorization rate (\%)} = [1 - (C_t/C_0)] \times 100 \quad (15)$$

Where, C_0 is the initial concentration (mg L^{-1}), and C_t is the concentration after the discharge treatment (mg L^{-1}).

And, the energy efficiency ($\frac{\text{mg}}{\text{kwh}}$) and the first-order rate constant (min^{-1}) for dye decolorization can be calculated by the following Equations 16-17 (Yamada et al., 2020).

$$\text{Energy efficiency} = \frac{C_0 \times v \times \frac{1}{100} \times \text{Decolorization rate}}{P \times t} \quad (16)$$

$$\text{Reaction rate constant} = \frac{-\ln(c_t / c_0)}{t} \quad (17)$$

Where, V is volume of the dye solution (L), P is consumed electric power (kW), and t is plasma treated time (min).

The state of helium flow is achieved by calculating the Reynolds number (R_e), which describes the stability of flow such as the stable laminar flow and the turbulent flow (Al-Rawaf et al., 2018). And this number can be calculated by the following formula (More et al., 2020):

$$R_e = 2.12 \times 10^{-2} \left(\frac{\rho}{\mu} \right) \times \left(\frac{Q}{r_D} \right) \quad (18)$$

Where, ρ is the fluid density (kg/m^3), μ is the viscosity (kg/m s), Q is the flow rate (m^3/s), and r_D is the inner diameter of the tube (m).

The quality of river water bodies that receive water produced by textile activities is not carried out by regular monitoring or control (Utami et al., 2019). So, it is necessary to analyze the impact of pollution and evaluate the role of wastewater treatment plants (WWTP) to determine the amount of pollution due to waste that has not been by the criteria (Utami et al., 2019). Furthermore, this assessment is needed to evaluate the action of the final treatment and wastewater unit and whether it is by criteria of quality standards. The result of this evaluation can be a recommendation for WWTP rehabilitation and optimization (Utami et al., 2019).

To assess the river's capacity for wastewater processing, it is essential to calculate the pollution load and compare it with established quality standards (Khalish et al., 2022). This evaluation helps identify the wastewater characteristics that require treatment and serves as the foundation for selecting the appropriate treatment units during wastewater treatment planning (Khalish et al., 2022). The quality standards used for this assessment are outlined in the Government Regulation No. 22 of 2021 concerning the Implementation of Environmental Protection and Management (Khalish et al., 2022). Equation 19 is used to calculate the concentration of the mixture

when water is discharged directly without any advanced treatment (Khalish et al., 2022).

$$C_c = \frac{Q_s C_s + Q_e C_e}{Q_s + Q_e} \quad (19)$$

Where, Q_s is the river discharge ($L s^{-1}$), Q_e is the effluent discharge ($L s^{-1}$), C_s is the river concentration (mg L^{-1}), C_e is the effluent concentration (mg L^{-1}), C_c is the mixing concentration (mg L^{-1}).

7 Techno-economic analysis

Although a considerable number of methods have been established for water and wastewater treatment, it is often believed that the operational cost of these advanced treatments exceeds the allocated budget for various treatment protocols, limiting their applicability at the industrial level (Mouele et al., 2021).

One obstacle currently impeding the transfer of plasma technology from the laboratory to the industry is the high cost of high-voltage power sources and total power generation (Okubo, 2021). NTP can be easily generated by applying a voltage between insulated electrodes. However, the energy consumption of NTP is a drawback for practical use (Okubo, 2021). This is also the case with various traditional techniques used in water purification processes. While they can treat water and wastewater, not all contaminants are removed (Mouele et al., 2021).

The circular economy aims to rely on sustainable resources, minimize toxic chemicals, and reduce waste through careful planning (MacArthur, 2013). Plasma technology can play a vital role in advancing the circular economy and achieving UNDP's SDGS goals (Figure 9) (MacArthur, 2013). Besides its eco-friendliness, plasma technology supports the production of products that replace or reduce harmful chemicals. For instance, in the textile industry, adopting plasma technology can contribute to achieving a circular economy (Reema et al., 2022; MacArthur, 2013).

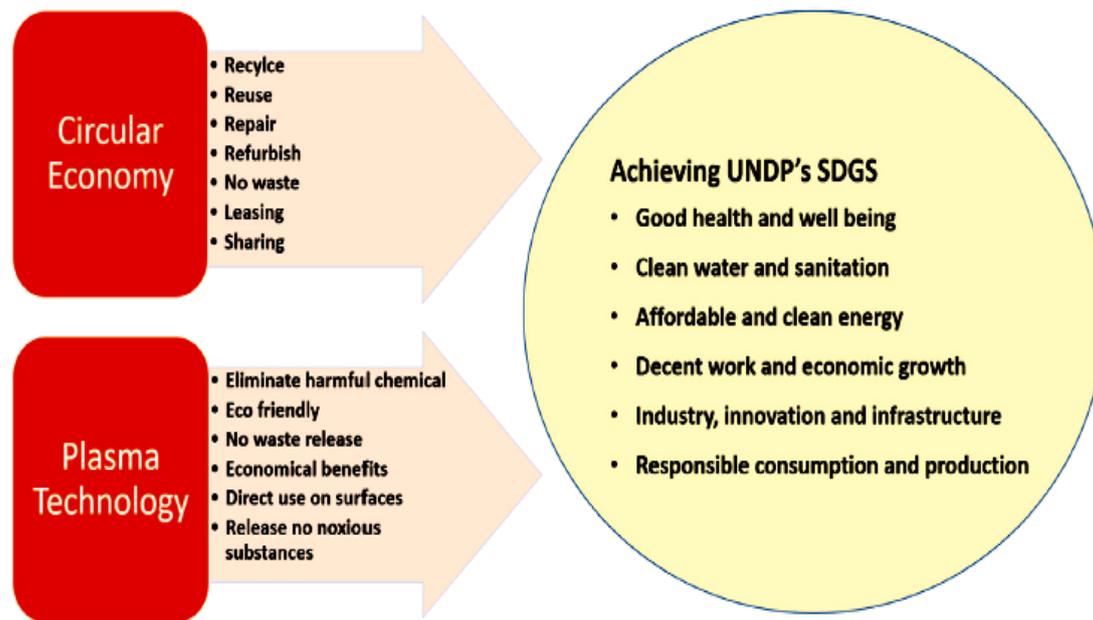


Figure 9 The relationship between the use of plasma technology and circular economy in achieving the UNDP's SDGs (MacArthur, 2013)

Plasma technology offers a solution to mitigate these risk factors. However, introducing plasma technology into the circular economy presents several challenges (Reema et al., 2022). The most effective approach is to commercialize it after extensive research. Establishing a connection between research and commercialization is crucial for a prosperous industrial future (Reema et al., 2022). Research should extend beyond laboratories and involve profit-driven enterprises on a larger scale (Reema et al., 2022).

Economic considerations may necessitate reactor replacement (Senthil Kumar et al., 2023). As the cost of renewable energy sources such as wind and solar continues to decline, nuclear power may become less competitive in the energy market (Senthil Kumar et al., 2023). In such a scenario, replacing an older reactor with a newer, more efficient design may emerge as the most cost-effective approach to sustaining a nuclear energy supply (Senthil Kumar et al., 2023).

8 Conclusion

Wastewater reuse poses challenges due to the associated costs in terms of both energy and finances. Effective management of these factors is essential to

harness the benefits of wastewater reuse for environmental preservation and resource conservation (Kusminah, 2018).

The review underscores the potential of NTP technology in assisting the textile industry. However, it acknowledges the hurdles that must be addressed. Various strategies exist for mitigating water pollution, but each approach comes with its own set of limitations. Despite ongoing efforts to enhance the cost-effectiveness of NTP technology, such as its integration with other systems, further research is imperative to fully leverage this technology for safeguarding the environment and human health, particularly in industries like textiles.

Although uncertainties persist in implementing plasma technology within these industries, research findings offer optimism, suggesting that these challenges are not insurmountable. Cold plasma stands out as a potentially groundbreaking and cost-effective technology with the capacity to address water pollution concerns.

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