PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0083766

The promise of non-thermal plasmas in addressing emerging environmental and health problems: present and future

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ABSTRACT

Non-thermal atmospheric pressure plasmas transform input electrical energy efficiently into reactive species, charged particles, and photons. This "activated gas" is being investigated as solutions for a range of environmental and health problems facing society today. In this perspectives review, we take a cursory look at a few of these societal problems and the reflected role that plasmas may play in charting the pathway to a solution buoyed by supporting research. Here we survey, the plasma-based opportunities in the removal of trace contaminants in water supporting methodologies such as water reuse which addresses scarcity and pollution, the opportunity posed by plasmas-based chemical depolymerization for plastics recycling, and the application of plasmas for food security, which includes sterilization of foodstuffs and the improvement of crop yield. Finally, we also included a short review on how plasmas may help control disease spread. In each case, the scope of the problem is presented along with the potential plasma-based solution.

I. PLASMAS AND THE ENVIRONMENT

A. The basis of a promising technologies

We stand at the precipice of a revolution in plasma science. Non-thermal atmospheric pressure plasmas (NTAPs) generated in regular air are poised to address a host of societal problems ranging from the environment to energy.[1] At the small-scale (liters), these discharges have demonstrated the capacity to remove pollutants in the air and water. Solutions to problems associated with agriculture such as the enhancement of produce lifetime and potentially the extension of the growing season via seed activation and preservation as well as the destruction of resistant bacteria and viruses on surfaces have all been demonstrated in the laboratory. Additionally, plasmas have shown signs of promise in the decomposition of carbon dioxide as well as plastics waste for recycling, thereby addressing a massive, emerging environmental crisis to come. These plasmas are currently also being explored to address a host of medical problems such as wound healing and tumor treatment.[2] In this perspectives article, we reflect on a range of environmental issues currently facing society today and survey plasma solutions contrasted with conventional methods where plasmas offer a compelling value proposition that warrants additional investigation. In many respects, the article serves as a map of societally impactful problems that have potential plasma solutions but remain nascent as underlying science questions remain unresolved. In large part, the value of plasma based solutions, particularly those that operate at atmospheric pressure is that they are non-equilibrium and thus offer high selectivity in plasma-driven chemical reactions and are inherently green—if powered by sustainable energy sources to produce the electricity. Before addressing various research areas, we briefly review the physics of atmospheric pressure plasmas and the key challenge inherent in all the potential applications—scaleup to the treatment volumes and surface areas that are of practical interest.

B. NTAPs

So, what then is a non-thermal atmospheric pressure plasma? These plasmas (also known as cold atmospheric plasmas, CAPs) are generated in typically one atmosphere of air and though not always but may also be operated in the presence of other inert feed gases to facilitate formation. Because a vacuum system is no longer required, in many respects implementation is simplified, and reaction rates can be higher with the increased mass activated by the plasma. In these plasmas, energy is imparted to electrons via high voltage pulses or some form of ballasting to circumvent the ionization instability which would otherwise lead to heavy particle heating and ultimately the transitioning into an arc.[3] In this case the Townsend avalanche is quenched prematurely before thermalization via collisions in the presence of the electric field can occur. In this respect the plasma is typically dynamic--actually a sort of ionization wave; that is a prebreakdown event. In many cases, the basis of NTAPs is a short-lived propagating ionization wave known as a streamer, not unlike a propagating lightning bolt. This ionization wave can propagate though any dielectric medium, essentially creating an ionization track in that medium. Figure 1 depicts the propagation of streamers underwater in the presence of injected oxygen bubbles.[4] A streamer is essentially the pre-breakdown phase of a discharge—existing during that time period before the interelectrode gap is breached. Because of mobility difference between ions and electrons charge separation occurs during the initiating avalanche leading to extreme electric field amplification at the streamer head or front. This field can exceed 10 times the sparking potential of the gap typically reaching several hundred kV per cm. This field drives ionization and allows the disturbance to propagate almost as if it has a mind of its own. Figure

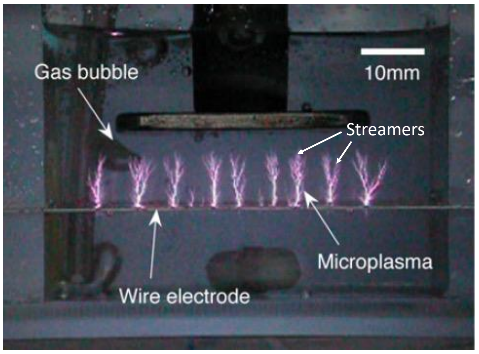


FIG. 1. Image of a streamer discharge propagating from a wire electrode underwater in the presence of oxygen bubbling.[4]

2 illustrates the spatial distribution of plasma particle and field parameters associated with a single positive streamer—one that emanates from an electrode excited by a positive voltage pulse. As can be seen in the figure, the high field at the streamer head drives ionization and the production of energetic electrons and photons. Because the field at the streamer head exceeds the applied field it propagates as if it has a mind of its own, tending to branch in places unexpected. Indeed, such branching is still now well understood though it may have some beneficial aspects in that it increases the volume through which the plasma interacts. Streamer propagation speeds in air are of order 10^7cm/s while an order of magnitude lower than this in water. The streamer has a diameter of order a mm and lengths that scale inversely with the neutral gas density. Researchers have found a relationship between streamer speed and size with the velocity scaling, with speed increasing with radius.[5] During its development as it propagates along in time and space, copious amounts of reactive species, charged particles and light emission are generated on timescales short compared to the thermalization time--typically ~ ns for prebreakdown processes in barrier discharges. The important point here is that it is the electrons that gain energy and activate the gas while the ions and heavy species remain relatively cool, typically near room temperature. The pre-breakdown event does not last long enough to create appreciable heating thus the ionization instability is avoided.[6] Indeed, the nonequilibrium nature of these plasmas provide a pathway to revolutionizing the way we do chemical processing--gone is the need for thermal reaction vats. Rather electrons directly drive reaction rates without the need to heat all the reactants to the reaction temperature. Indeed,

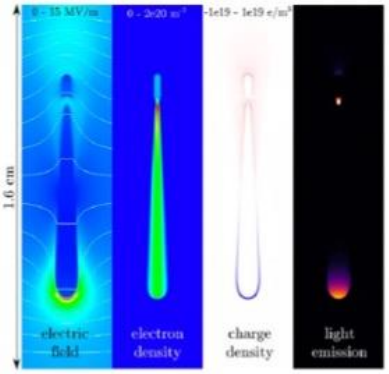


FIG 2. Simulation of a positive streamer propagating downward (exciting voltage 32 kV in 1 ATM)[5]

the key to non-thermal plasmas is they afford one the opportunity to exquisitely control reactivity through bond breaking and electron donation--literally tailoring the reaction pathway. In this respect there is direct communication between input power and the reaction by way of the coupling of electron energy to input process mass flow. Here if the plasma is made of air, then oxidants such as ozone, OH, and hydrogen peroxide to name a few form. Additionally reduction agents such as hydrogen and if in the presence of water, solvated electrons—the strongest reduction agent known to chemistry, are also formed. Therefore oxidation and reduction can be directly driven by plasmas and to some extent selectively controlled for applications ranging from chemical synthesis to decomposition. Here, the plasma phase can be thought of as extreme radical chemistry.

In general, the plasma sources currently being explored for the wide variety of environmental applications include: 1) the dielectric barrier discharge, 2) the pulsed streamer discharge and 3) the pulsed corona discharge. By far the most widely explored discharge type is the dielectric barrier discharge. In fact, the basis of ozone generation industrially for water treatment applications is the dielectric barrier discharge explored by Siemens dating back to 1854. Figure 3 illustrates the basic operation of a dielectric barrier discharge.[2] Shown in Fig. 3 is a parallel plate geometry with electrodes covered with a dielectric(the barrier) which operates in regular air under ambient temperature and pressure conditions. If AC voltage is applied, typically around 20-30 kV at a frequency of 10s of kHz the gap will breakdown creating a host of reactive oxygen species—including ozone and reactive nitrogen species. Charge from the breakdown plasma deposits on the electrodes eventually to the level where the associated electric field reduces the total electric field below the sparking

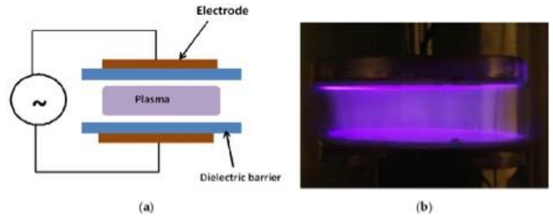


FIG. 3. Schematic depiction of dielectric barrier discharge along with photograph of operating discharge.[2]

potential of the gap and the discharge goes out. The process repeats itself every cycle with the dielectric barrier restricting current growth and lifetime (ns's) such that the thermal instability is avoided—the heavy species (ambient air) does not heat up because the discharge is not on long enough. This approach is an efficient way to produce reactive species. While Fig. 3 shows a relatively diffuse discharge, in actually the discharge is actually filamentary, consisting of a multitude of short-lived streamers. A pulsed corona discharge is more or less a pulsed version of a typical high voltage corona discharge. Recall a corona discharge forms on high curvature surfaces at high voltage such as power line wires owing to the high local electric that results. This field locally ionizes the gas near the electrode. Current continuity is due to charge drift that completes the circuit. If one increases the voltage and thus electric field , the active region near the electrode will expand until it reaches the return electrode resulting in the formation of a thermal spark. To avoid this issue and realize higher discharge currents resulting in the production of more reactive species, one can apply a short duration, fast rise time high voltage pulse. The ionization wave propagates at high speed upon application of the pulse producing many reactive species as the current builds. The discharge terminates owing to the short duration of the voltage pulse, which prevents the discharge from bridging the gap and producing a thermal spark. Here pulse lifetimes are of order 100s ns for typical interelectrode spacings with ionization wave speeds of order 10^8 cm/s in air.[7]

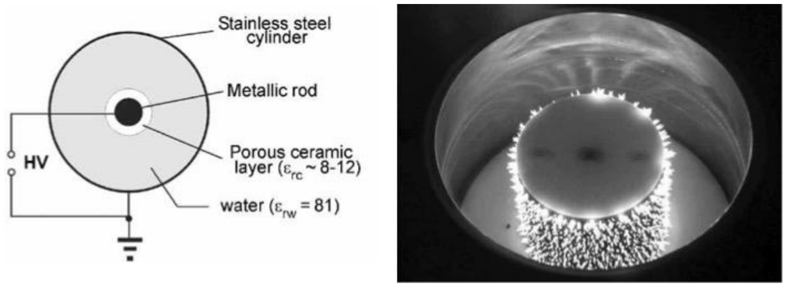


FIG. 4. Pulsed corona discharge geometry and operation.[8](Adopted from Lukes, 2009)

Figure 4 illustrates a pulsed discharge operating underwater.[8] As can be seen here it has the appearance of a multitude of streamers. The voltage pulse terminates before the discharge can cross the gap. Such discharges are particularly effective in producing reactive species in air as well as in liquid water.

Still another well studied nonthermal plasma source is the plasma jet. The plasma jet is characterized by its luminous plume emanating from a microdischarge plasma in the presence of gas flow, spanning cm's away from the source. The plasma jet can be configured in a number of geometries as can be seen in Fig. 5 but its key attribute is that its plume or jet of localized light emission and reactive species, making the device applicable for host of medical applications requiring precision localized treatments.[9-11] The jet is formed by the production of an ionization wave—a streamer-- launched by the powered electrode which then propagates along the channel of gas flowing out of the device as depicted in Fig.5. It can be thought of as guided streamer where here the ionization wave propagates in the channel of helium feed gas.

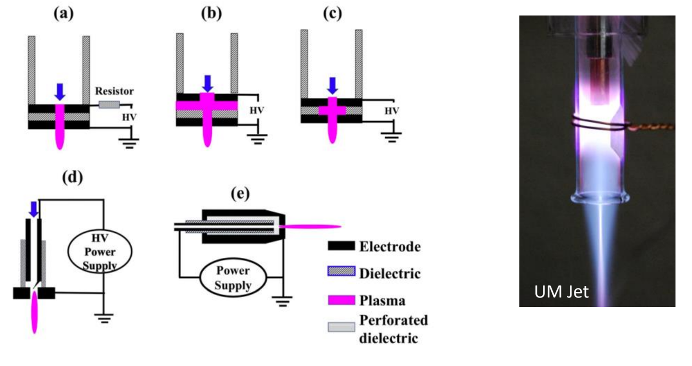


FIG.5. Plasma jet implementations[11] (adopted from Lu, 2021) and photograph of operating jet from PSTL UM lab-B. Yee)

This propagation or apparent confinement and stability occurs because the ionization coefficient is considerably higher in the helium jet in comparison to the surrounding air at the low reduced electric field for which the jets are formed it. This guided propagation results in the formation of a directed jet without branching.[12] It turns out that for discharge tubes greater than about a mm, the jet is actually hollow, largely due to the fact that plasma breakdown starts near the walls of the dielectric tube. Density and field are maximum at the dielectric wall near the exit plane thus this annular region is the source of the streamer propagation front. The jet's length and stability is related to the flow regime of the exiting gas—with longer, more stable jets favoring laminar flow over extended distance from the exit of the device. For positively applied voltages, a cathode seeking streamer is formed and the jet itself in this case can be thought of as a discharge between plasma at the exit and ground. High speed imaging of the ionization wave suggests a single solitary "plasma bullet" ionization wave, but owing to the high field at the streamer head, the discharge is most luminous there as compared with the streamer tail.[13,14] In fact, there is an ionization trail extended from the tube exit to the tip of the jet. It has also been suggested that the preionization needed for streamer propagation was due to photoionization ahead of the front. As the ionization wave propagates along the exiting gas it produces copious amounts of reactive species. While the jet can operate on argon, it typically is most stable when operated with helium as the feed gas which makes it less susceptible to the ionization instability owning to helium's high ionization potential and high thermal conductivity. As can be seen in all discharge cases described here the plasma contact area is very limited typically measured in square mm for each streamer or microdischarge footprint—which represents a key challenge question—how do we practically scale the sources up for practical applications? This is a source of intense research as will be touched upon in later subsections.

C. The challenge

The opportunities presented by non-thermal atmospheric pressure plasmas are indeed many, but the promise can only be realized if scaleup is possible. By scaleup we refer to plasma processing at volume scales and over time intervals that are typical of industrial requirements. For example, water is treated at wastewater plants in the millions of gallons per day. In this particular case, the input power, the introduction of reactive species, and treatment time must all be commensurate with typical treatment flows at power levels that are not prohibitive for practical implementation. So, then the challenge to environmental plasma applications is to scale the plasma up in spatial extent with required reactivity. The impediment to scaleup that sums up a key struggle in the field of NTAPs is inherent to the nature of the way these plasmas are formed. At one atmosphere the pre-ionization streamer is filamentary with diameters on the order of a fraction of a mm. This means that contact area with the process mass is limited. And because the reactive species are introduced at these contact points for envisioned applications, treatment rates for large volumes become quite challenging. The community is innovating in this area through the use of geometry and in-volume treatment methods to address this very problem. As such scale up is application specific, various approaches will be posed in the sections that follow.

D. The Scope

At this point in human history, we face an unprecedented array of environmental challenges affecting the quality of life. Here we survey the opportunities posed by non-thermal atmospheric pressure plasmas to address societally impactful problems such as environmental threats to air and drinking water, to provide resilience in agriculture in light of climate change, provide a novel and efficient method of chemical processing, to close the plastics pollution loop, and to address the threat posed by resistant bacteria and viruses. Alongside the societal issues that we face today that are also identified here, we present shortcoming or gaps in conventional approaches and the opportunities afforded by plasma solutions currently being investigated along with challenges to realization and prospects for the future. While this is not meant to be a comprehensive review, our goal is to inform the reader of the exciting areas of research in plasma science and the promises such research holds if realized for the betterment of society.

II. THE CHALLENGE OF CLEAN WATER FOR ALL

In 2010, the UN declared access to clean drinking water and sanitation a human right. Indeed, access to clean water is now a part of the UN's 8 Millennium goals aimed at meeting the needs of the poorest.[15] Currently nearly 800 million people lack access to clean drinking water and about 2 billion with access to water services but with water contaminated with human waste.[16] The urgency of this societal problem has prompted the National Academies of Engineering to designate "clean water for all" a National Academies Grand Challenge. Inadequate wastewater treatment is a key contributor to water pollution. In this case, the contribution to water contamination from human and animal waste is particularly problematic leading to millions of deaths each year from waterborne illnesses in underdeveloped regions especially. Indeed, as illustrated in Fig. 6, the number of people dying each year from unsafe water is staggering, dwarfing natural disasters and conflict-related deaths combined.[17] Note the persistence in underdeveloped and developing countries. This problem is exacerbated as population density increases in a given area that lacks adequate wastewater treatment capacity. The occurrence of emerging pathogens in water is another issue that is aggravated by overpopulation and over industrialization.[18] These emergent pathogens arise for a number of reasons. For example, population shifts to new regions driven by climate shifts and deforestation can lead to new exposure modalities. The implementation of new technologies regarding water management such as dams and irrigation, wastewater treatment and discharge practices and mismanagement of agricultural waste also contribute to the introduction of emerging pathogens. Additionally, the emergence or reemergence of

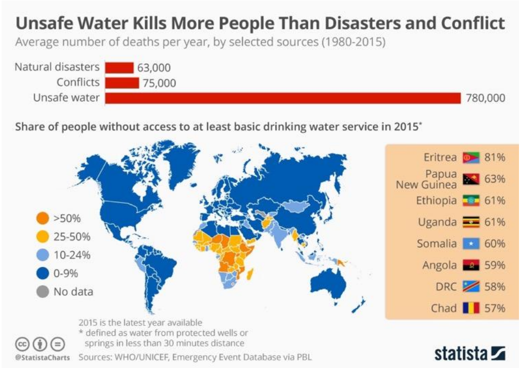


FIG. 6. Scale of unsafe drinking water worldwide.[17]

waterborne diseases can result from large-scale changes in human behavior and conditions, for example such as humanitarian crises, the increase in the population of those with underlying health conditions, and the ease of human travel worldwide. Also, pollution derived from discharges into freshwater bodies from industrial operations such as textile mills and chemical processing plants play a role in limiting availability. Inadequate removal of industrial toxins in drinking water can lead to long term health problems such as cancer. Another driver limiting drinking water availability is associated with misuse of scientific advances in development of antibiotics, herbicides and insecticides. Overuse of these chemicals can also lead to new and emerging pathogens. Beyond pathogens, pharmaceuticals, dispensed in waste streams can further lead to resistant microbes thus exacerbating the pollution effect and further highlighting the need for advanced treatment methods and water management policies.[19] This problem is further compounded by climate change which is expected to give rise to water scarcity in certain regions of the world.[20] In the underdeveloped world, developing countries, and developed countries, addressing water scarcity derived from climate change will be a major challenge in the coming decades. This scarcity not only impacts the quantity of water available for drinking but also impacts food production as over 80 percent of all freshwater draws in the US for consumptive use (removed from the environment e.g. metabolic incorporation by plants or livestock) is associated with agriculture.[21] From a practical standpoint, those advanced treatment solutions ultimately selected to address this crisis, the technologies must be implementable at scale and compatible with resources available.

A. A potential solution: Water reuse and the need for advanced technologies

Regardless of the development level of a particular region, scarcity will be a common theme in those regions experiencing reduced precipitation owing to climate change and population growth leading to an imbalance between withdrawals and replenishment by natural processes. Indeed, the worldwide population is expected to reach 9.7 billion by 2050.[22] Pollution also limits water availability. Even at low concentration levels some forms of contaminants can impact human health. Compounds such as PFAS (per- and polyfluoroalkyl substances) and 1,4 dioxane, carcinogens, are now pervasive in ground water. Indeed, over 1/5 of all drinking water supplies in the US contain traces of 1,4 dioxane. This compound has a half-life in the groundwater of 2-5 years.[23] On the other hand, PFAS compounds do not degrade readily in the environment and currently its environmental half-life is unknown. One such long lived species is shown in Fig. 7.[24] These so-called contaminants of emerging concern in drinking water have led to the recognition of the need for advanced treatment technologies even in those regions where water is readily available. Such technologies are enabling in that they also have the potential to support the realization of a methodology that directly addresses scarcity--water reuse. Water reuse is a water management approach whose associated technologies address both scarcity and the occurrence of contaminants of emerging concern in fresh water sources. Water reuse involves the reclamation and reuse of wastewater effluent using additional treatment processes to make the resulting recycled water available for a range of applications, both potable and nonpotable. Here, while normally water is recycled after treated wastewater discharge is processed in the environment, reuse usually refers specifically to bypassing the environmental step and typically sending the effluent directly to an advanced treatment plant. Figure 8 illustrates the water reuse approach.[25] Here water is treated for "fit-for-purpose" where the treatment trains utilized depends on the requirements for the end water use, for example process water for power plants. This approach is resilient to drought conditions and contamination via pollutants from

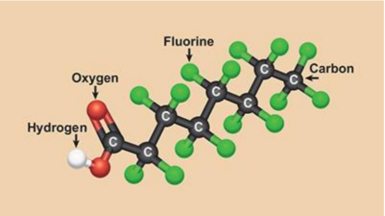


FIG. 7. PFAS compound PFOA, a persistent environmental contaminant used as a surfactant in firefighting foam and carpet care products---a component of Teflon.[24]

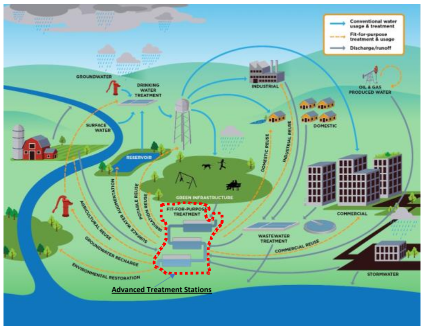
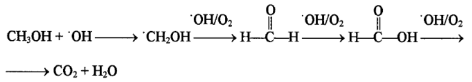


FIG. 8. Water reuse—source water is treated via advanced methods to the degree to meet end use requirements (courtesy of US EPA).[25]

external sources. Additionally, it removes the prospect of wastewater discharge damaging habitats and addresses supply chain concerns. To facilitate reuse, extensive treatment is required in particular to remove contaminants and pathogens. These very same advanced technologies will be needed for conventional water treatment plants as well as regulations for contaminants of emerging concern are increasingly regulated with actual maximum concentration levels levied by utilities by State and Federal environmental protection agencies. States such as California require > 10 log reduction in a few contaminants in reuse water--a challenging target for current technologies. Some of these contaminants enter the drinking water system owing to aging infrastructure issues such as line breaks, biofilm formation and leaching, which can lead to the intrusion of microbial, and chemical contaminants.[26] Contaminants of particular concern include 1,4 dioxane and PFAS along with pathogens such as cryptosporidium, Legionella, and Escherichia Coli. Urgency is highlighted by the potential for bacteria in wastewater to gene exchange giving rise to antibiotic resistance.[27] These challenges illustrate the need for robust treatment technologies that can handle changes to water quality over a broad spectrum.

B. The approach--conventional methods

Conventional water treatment methods date back nearly 100 years and at its core features filtration to remove particles and disinfection. Such technologies do not directly address aforementioned challenges to reusing water for potable water applications or even the removal of contaminants of emerging concern that are not present in waterways and groundwater. Advanced methods are certainly required at this point. Driving the infusion of advanced technologies is regulatory pressure which defines which chemicals to monitor and the associated maximum concentration level. To meet these requirements, new technologies and diagnostics are integrated into treatment trains.[28,29] In support of water reuse, advanced oxidation processes (AOPs) have been identified as a potential solution in combination with reverse osmosis (RO). Advanced oxidation acts as the second barrier for those contaminants that escape the RO filtration. Advanced oxidation methods involve those chemical processes aimed at the production of OH radicals in solution.[30,31] OH in the presence of oxygen mineralizes organic contaminants in solution into CO_2, water and inorganic salts. The process involves steps such as hydrogen abstraction and addition. The decomposition of methanol via the OH radical attack is shown in Fig. 9. Attack of more complex molecules leads to an even more complex chain of events. The chemical pathways which lead to intermediate compound formation along the way to mineralization are still not well understood and are currently under study.[32] Indeed, one can argue that such complex decomposition pathways could potentially be explored using machine learning algorithms, which may enable the development of a predictive tool to assess decay routes. Intermediate radicals have been summarized elsewhere.[30,31,33,34] AOPs require precursor chemicals to drive OH production in solution such as ozone or hydrogen peroxide.



$$CH_3OH + {}^{\cdot}OH \longrightarrow {}^{\cdot}CH_2OH \xrightarrow{{}^{\cdot}OH/O_2} H\text{—}\overset{\overset{\displaystyle O}{\|}}{C}\text{—}H \xrightarrow{{}^{\cdot}OH/O_2} H\text{—}\overset{\overset{\displaystyle O}{\|}}{C}\text{—}OH \xrightarrow{{}^{\cdot}OH/O_2} CO_2 + H_2O$$

FIG. 9. Decomposition of Methanol via hydroxyl radical attack[33]

While AOPs are effective against a range of most organic compounds, it is not as effective against fluorinated organic compounds such as PFAS.[35] These compounds have now proliferated in the watershed owing to its mobility and recalcitrant nature. Such compounds may be removed from drinking water via reverse osmosis or activated carbon. Both processes transfer the contaminant from the water stream into a secondary waste stream--concentrate in the case of RO and loaded carbon in the case of activated carbon--which all require disposal. Advanced reduction methods generate reducing agents such as the solvated electron to degrade fluorinated compounds in liquid water.[36] There is therefore a need for scalable, cost-effective methods that are agile enough to address future treatment needs as they arise but also adoptable to regions of reduced infrastructure where in such cases the need is great.

C. Plasmas --gaps in understanding, the need, future prospects

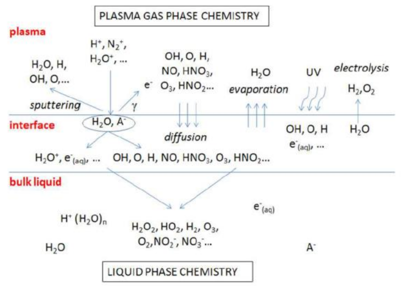


FIG. 10. Complex processes occurring at interface of plasma with liquid water.[39]

The interaction between non-thermal plasma and liquid water gives rise to the introduction of a host of reactive species into the water (see Fig. 10). Indeed, the interaction is also physical in that plasma is a source of heat, ultraviolet light, energetic electrons, metastables, ultrasound and shockwaves.[37-39] These physical processes are all a form of advanced oxidation methods. In this case however, the processes are all essentially occurring simultaneously in contrast to conventional advanced oxidation where a single process is typically feature such as the combination of ozone and peroxide. This potent array of active ingredients has shown efficacy in the removal of a host of contaminants including fluorinated compounds such as PFAS at the laboratory demonstration scale. On the other hand, scaling up this technology for larger scale use has proven to be challenging. The challenge lies in the very nature of the plasma liquid interaction at atmospheric pressure--the condition required for practical implementation in water treatment facilities for reasonable reaction rates and overall reduced system complexity and cost. NTAPs can facilitate the removal of contaminants in water via 2 routes: 1) physical destruction via bond breaking (e.g. energetic electrons) or 2) chemically via advanced reduction or advanced oxidation. These processes are achieved by bringing together water with plasma directly or indirectly. Indeed, the plasma can be generated directly in water via the application of a high voltage pulse which gives rise to the formation of a propagating streamer. A streamer is a type of ionization wave that can propagate through a dielectric medium driven largely by an intense electric field that forms from charge separation as a pre breakdown process. This field at the head or ionization wave front of the disturbance gives rise to fields 10 x the applied field. Along the trail of ionization in water are copious amounts of OH radicals, solvated electrons, UV light and shock waves associated with a collapsing ionization channel. Such a discharge requires high voltage to breakdown water and the energy cost is of order a Joule per pulse. It has been theorized that the streamer actually propagates via microbubbles either present in the water previously or those generated by the streamer head itself. One can also generate a plasma via independent means and place the discharge in contact with the surface of the water. This method is typical of dielectric barrier discharges (DBDs) and plasma jets. DBDs are a clever approach to producing non-thermal plasma at high pressure, avoiding the formation of an arc. If one considers voltage applied between two electrodes with at least one covered in a dielectric, at the sparking potential a discharge will form but it quickly extinguishes when the charge deposited on the dielectric reduces the field below that which can sustain the discharge. Application of a sinusoidal voltage thus produces periodic breakdown spikes which are the source of active particles to interact with the water. The surface interaction gives rise to the production of reactive species. Water treatment featuring this approach relies on diffusion to propagate the reactive species through the volume and thus treatment times can be slow. Another drawback of this type of discharge which also plagues the underwater streamer variants is the limited contact area that the plasma makes with liquid. At atmospheric pressure, the mean free path of electrons is of order microns and thus the physical size of the discharge itself in contact with water is small--usually filamentary with small sub-mm to mm footprints. The limited contact area poses a significant challenge to scale up. One way around this problem is to produce the plasma in bubbles monodisperse throughout the volume. In this way, reactive species can be transported throughout the volume provided their active lifetime is long enough in the gas phase. Past studies have shown that in general the most efficient reactors feature those configurations where plasma is exposed to thin layers of water.[40] The result is directly a consequence of the fact that the treatment is a surface process where active species have to diffuse into the bulk for treatment. In this respect, embodiments of reactors that have been scaled up feature the application of a multitude of microdischarges attached to a thin layer of water flowing underneath.

D. Applications now

Despite the inherent scaling problem with high pressure plasmas, there are geometrical approaches can lead to de facto large area or in-volume treatment. Indeed, plasma-based water treatment technology in which geometry is manipulated to achieve greater contact has been explored commercially. AQUAPURE®, one of the first companies to attempt to commercialize a plasma reactor, fielded an air plasma discharge where the water treatment volume was disposed into a thin layer which in conveyer belt fashion flowed under a multitude of corona discharges. This geometry maximized surface coverage by simply utilizing a large number of discharge applicators. Additionally, because the treatment layer was thin, the plasma dose delivered to the water as it flowed under the gauntlet of discharges was enough to treat that volume to the targeted maximum allowable concentration levels. The device could treat up to 4 liters per minute. This apparatus was actually piloted multiple industrial settings. DMax Plasma LLC is currently developing a reactor for the removal of PFAS compounds.[41] The reactor also features a multitude of pulsed corona discharges to address the contact area

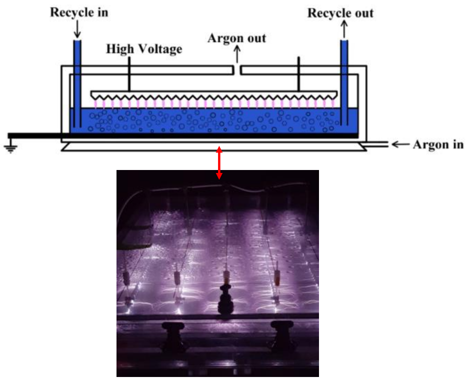


FIG. 11. Multi-applicator argon gas discharge for the treatment of PFAS in water.[41]

problem and treat the liquid in batches. The contaminated water to be treated is disposed to a thin layer. The surface contacting discharges are ideal in that the PFAS species are hydrophobic and thus portions of the molecule are present at the surface and thus will come in direct contact with the plasma. This discharge features argon gas as can be seen in Fig. 11 which maximizes the solvated electron generation by way of increased plasma density. The water is batch processed in an argon gas atmosphere contained within the reactor vessel, thus mitigating the need for continuous argon feed. Figure 12 shows the efficacy of the reactor at removing PFAS compounds from filtered ground water derived from an Air Force remediation site with efficiencies ranging between 9 and 31 kWh/m^3. Here cycle number refers to the number of times the water is re-introduced to the reactor. The behavior has an exponential decay-like form suggesting a first order-like rate for decomposition.

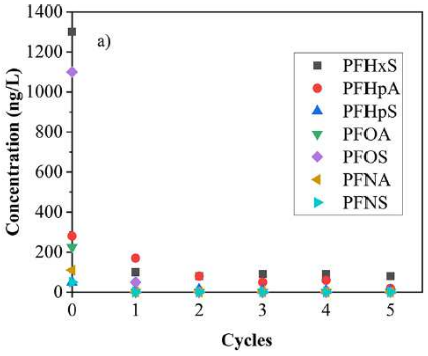


FIG. 12. Decomposition of a range of PFAS contaminants as a function of cycles through the reactor.[41]

Another approach to scale up is the so-called pulse plasma discharge with dielectric barriers. This reactor, rather than treating thin sheets of water, disposes water flow through standard tubing into thin streams via a shower head applicator. In this manner, the effort addresses the surface area plasma contact problem that plagues plasma-liquid applicator approaches.[42,43] The water streams act as leaky dielectric barriers. The reactor is shown in Fig. 13. The plasma then interacts with these

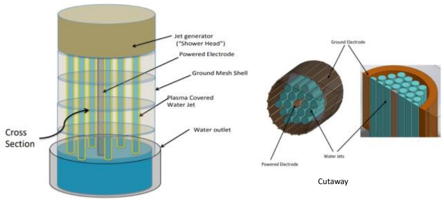


FIG. 13. Cutaway of packed bed plasma water reactor. Plasma flows along water streams as surface ionization waves.[43]

high surface areas to volume structures via ionized gas formation in the interstitial spaces between the streams and via surface ionization waves that propagate along the water streams themselves. The discharge is generated in regular air. The approach is scalable from the standpoint that one can utilize a multitude of applicators in parallel to attain higher flow rates. This reactor is also being investigated for the treatment of PFAS compounds in groundwater derived from industrial operations some time ago.[43] Here again, it is postulated that it's the deposition of solvated electrons that facilitates the decomposition of the fluorinated compounds. Figure 14 illustrates device operation on PFOA spiked deionized water with starting concentration of 5.7 ppm at an efficiency of around 45 kWh/m3, which falls within the median range of efficiency for conventional advanced oxidation methods such as UV-peroxide.[43]

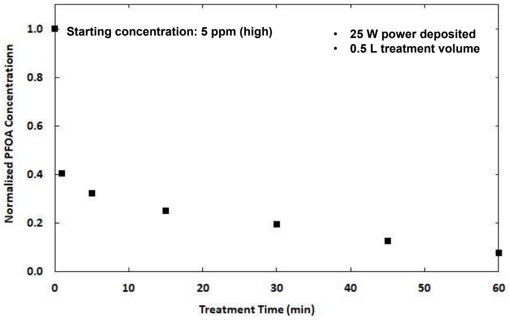


FIG. 14. Forever chemical PFOA removal via batch processing with packed bed reactor.[43]

E. Future prospects

Plasma based water purification offers a compelling solution to a host of water quality problems. The physical mechanisms of decomposition are fairly nonselective and thus indiscriminate, making it ideal to address a range of contaminants including those accidentally introduced or those that are currently emerging. This attribute is ideal in that it addresses unregulated chemicals present in the water as well as those that become controlled without the need for modification of the reactor. Such technology makes the water treatment trains also resilient against accidental spills. The technology has the potential to eliminate the need for consumables as well as complement conventional methods such as RO by treating the reject-water, which otherwise is a waste disposal issue. The exciting aspect of plasma-based systems is that it addresses some water quality problems whose treatment solutions are too expensive to carry out conventionally and thus provides an opportunity for plasma technology. The PFAS problem is just one such example. However, scaleup and energy efficiency are parameters that need to be fully quantified before there is large scale adoption. Herein lies plasma physics opportunities in the areas of improved understanding of the plasma liquid interface including understanding and ultimately exploiting poorly understood phenomena such as plasmas self-organization which has been shown to affect not only contact surface area but also mass transport via induced flow.[44] From a chemical perspective, there is still much to be learned regarding the decomposition pathways driven by plasmas as it is critical to understand the degree of mineralization and the level of toxicity of intermediates formed during processing. This particular problem is rich in molecular, physical and organic chemistry. Machine learning in combination with the reactor is one possible approach to predicting and controlling decomposition particularly of the smaller chains that form. In this respect, with this knowledge, plasmas as a treatment tool may hold the key to how we provide clean water to the world including large-scale water reuse. The enterprise includes engineering and physics to make this promising technology a reality.

III. THE PROBLEM WITH PLASTICS

Plastics waste stand as another looming environmental problem facing society today. The amount of plastic produced in the world is growing exponentially. As an example, in 2017, there are on the order of 1 million plastic bottles purchased each minute worldwide.[45] While this production supports societal needs, most of the plastics produced at the end of life become waste. Projections suggest that the amount of plastic waste will top 12 billion metric tons by

2050. For reference, to date approximately 8.3 billion tons of plastics has been produced worldwide in total. Only 10% of this waste globally is recycled—which means that this waste is accumulating, unsustainably in the environment. Indeed, 79 percent of all plastics ever produced is in landfills. And in aggregate approximately 90 percent of all plastics ever made are not recycled.[46] Figure 15 graphically depicts the accumulating plastics waste problem.

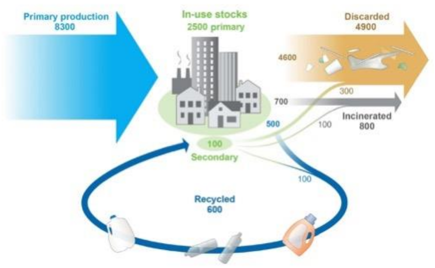


FIG. 15. World-wide plastics production and subsequent fate with numbers indicating mass in million metric tons.[46]

This accumulation is giving rise to a whole host of environmental problems ranging from destruction of marine habitats to the release of chemicals of emerging concern, particularly those related to carcinogenic additives used to make plastics more durable. Additionally, this material in the form of mm to microsized particles known as microplastics are finding their way into the human body. These particles come from either those products that use microplastics as an ingredient such as cosmetics and clothing items such as microfibers from synthetic fabrics or from crumbling due to the physiochemical degradation of once larger particles in the environment. It is estimated that in America, humans ingest up to 52,000 particles per year. These particles can enter the body through the ingestion of contaminated food and drinks. It should be noted that an additional 9000 per year is consumed if one's drinking water is derived exclusively from bottled water. On the other hand, those who drink from the tap consume about 4000 particles per year.[47] These particles can also be ingested through inhalation. Recently it was reported that inhalation of plastics dust derived from microfibers in the household can be significantly larger than for example by the uptake from shellfish. Indeed, the household source has been estimated to contribute to over 60,000 particles per year.[48] The accumulation of plastics in the body is problematic in that various plastics have varying degrees of toxicity with can affect the immune system as well as release toxins into the body such as heavy metals and PCBs (polychlorinated biphenyls).[49] Additionally, colored plastics often contain very toxic pigments which can leach into the water or soil.[50]

While the plastics pose a host of environmental issues, the exponential growth in plastics production is also not sustainable as it is derived from a limited resource—fossil fuels such as ethylene and propylene.[51] It should also be pointed out that in the environment driven by photochemical degradation, plastics emit gases such as ethylene and methane. These are global warming gases and thus as plastics proliferate in the environment as waste, their contribution to climate change can be expected to increase as well thus compounding the problem of plastics.[52]

The production of plastics is a thermal process, and, in this regard, its production has a large carbon footprint. In the environment, plastics decay at an alarmingly slow rate. For example, the degradation half-life for a simple plastic bottle is 58 years and the degradation of plastic pipe is a whopping 1200 years! Degradation rates for different types of plastics is shown in Fig. 16.[53] The ultimate solution to this crisis is simply to cease the production of new plastics and harvest the waste plastic.[54] By depolymerization of the recovered plastic waste, one can in principle produce monomers and precursors necessary to make new plastics thus closing the cycle.

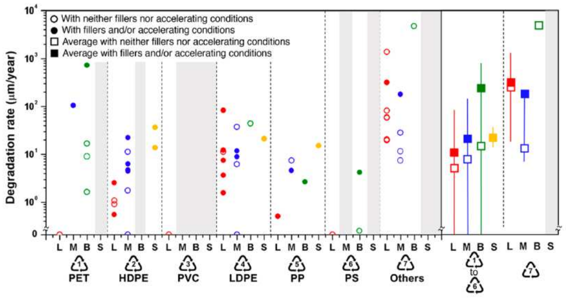


FIG. 16. Degradation rates for different types of plastics, where letters designate degradation environment. L refers to landfill, M-marine, B-biological degradation and S-sunlight.[53]

Currently, plastic waste is generally treated in one of four ways. 1) Primary recycling where well separated materials are simply reheated and then molded into another product. Typically, with each cycle, the material loses some mechanical properties of the earlier generation. 2) Secondary recycling involves the integration of plastics waste into other products, typically of lower value after the plastics stock has been reheated, extruded and milled. 3) Tertiary recycling is most desirable in that it uses chemical processes to convert the plastics to monomers leading to the production of higher value products or fuels. Quaternary processing involves either incineration or deposition into a landfill.[54]

A. Challenges in plastics recycling

A key challenge in plastics recycling is the identification of the specific plastics type and appropriate separation. In general, there are 6 basic plastic types: 1) Polyethylene Terephthalate (PET) used in clear plastic bottles, High Density Polyethylene (HDPE) used in products such as white milk containers, Polyvinyl Chloride (PVC) used in piping, Low Density Polyethyelene (LDPE) used in plastic bags, Polypropylene (PP), used for bottle caps, and Polystyrene (PS), (styrofoam) used in cups and packaging material. The goal of recycling is to reduce a given plastic to simple monomers--the simple base molecular building block of the plastic. These can then be reconstituted to make new plastic of the same type or even processed to higher value products. The presence of other plastics in the process chamber can therefore have deleterious effects. Indeed, contamination can render a batch useless. For example, a single bottle of PVC in a batch of PET plastic bottles can render the entire batch unusable--a daunting challenge of recycling. This is due to the fact that at PET processing temperatures, the PVC will char, contaminating the lot.[50] Mixing PP with HDPE for example can also greatly alter mechanical properties of the recycled material leading to for example increased brittleness.[54] Effective recycling is therefore highly dependent on the purity of the batch of plastics to be processed. Sorting is therefore a critically important step and challenge in plastics recycling. So accurate sorting is key to plastics recycling. For any practical recycling facility, this sorting must be done on the fly to facilitate separation upon entry especially given the sheer volume of plastics waste. Laser induced breakdown offers one possibility for on the fly identification of plastics material type. Here a laser vaporizes a portion of the plastics sample. The spectral emission from the plasma yields insight into the signature of a given plastic.[50,54,55]

Electrostatic separation is also possible with plastic powder feedstock for example. Because dissimilar plastics charge up differently upon rubbing, it is possible to separate plastics based on their triboelectric properties as well. Charging can be achieved in a fluidized bed. Particles can then be separated as they fall under gravity through an applied electric field, orthogonally oriented so as to deflect particles to their respective bins. The separation of PVC and PET at high recovery percentages has been realized with this method.[56,57]

Selective flotation is also a means to separate particles. Here one relies on relative differences in the hydrophilicity of the plastics. If one plastic is made more hydrophilic than the other, then one can use bubbles for example to essentially float out the more hydrophobic species. The process is dependent on surface chemistry as well as particle geometry. It has shown promise for the separation of PET and PVC.[58]

Once the plastics are separated, they must be depolymerized; that is, they must be broken down to the basic building blocks of the polymer chain. For example, cellulose, a long chain molecule, can be broken down into glucose units, which make up the molecule: $(C_6H_{10}O_5)_n$-->$n(C_6H_{10}O_5)$, an important reaction for energy production in living organisms. And in the case of polyethylene, the building blocks are the ethylene molecule chained together (C_2H_4). In the case of plastics, the building block chemicals or monomers can be later used as feedstock to construct new plastics with similar properties or entirely different types of plastics. The key is depolymerization. Depolymerization of PET plastics can be carried out via hydrolysis, which can be carried out under acidic, basic or neutral conditions. In hydrolysis, a water molecule essentially cleaves the bond of the PET molecule leaving dimethyl terephthalate (DMT) and ethylene glycol (EG).[59] The desirable attribute of this reaction is that the resulting ethylene glycol and dimethyl terephthalate resulting from depolymerization (or terephthalic acid) can be used to make new PET plastic. Other plastics such as polyethylene are not as straightforward to depolymerize. The difficulty lies in breaking up this molecule to low molecular weight products that can be used to produce new plastics. Currently, catalysts are being explored to facilitate controlled disassembly of this molecule and others at reduced temperatures and high selectivity. The key goal here is to cleave the molecule at the catalytic surface to generate a monomer unit. Polymers can contain up to a million monomer subunits and thus getting the polymer in contact with the catalyst as well as preserving the catalyst surface conditions are also important considerations. Carbon deposition on catalyst surface for example can reduce the number of active sites for depolymerization. These challenges remain an active area of research.[60]

B. Plasmas and the Plastics Problem

The plastics problem stands as an opportunity for plasma engineering. Already, plasma pyrolysis has been investigated as an approach to address the plastics problem. In plasma pyrolysis, a thermal plasma arc jet with temperatures up to 10,000 K is used to vaporize and dissociates molecules making up the plastic waste, which is typically injected into the reactor in the form of powder or pellets. The process is not sensitive to plastic type and thus waste with varying types of plastics compositions can be processed at once. Figure 17 depicts a plasma gasification system. Here plastic aggregate is pretreated before being introduced to the thermal arc shown in the process tank struck between two graphite electrodes.[61,62] During the process these molecules are partially oxidized by the high

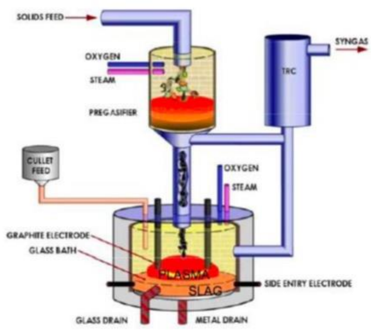


FIG. 17. Plasma pyrolysis reactor for gasification of plastics.[61]

temperature (>1500 C) plasma gas. By partial oxidation, we refer to the stoichiometrically incomplete mixture of oxygen and organic compounds such that the by-product is not simply carbon dioxide and water but rather carbon monoxide and hydrogen with at lower concentrations water, carbon dioxide and other trace gases dependent on original composition of the plastic waste.[63,64] As such, plasma pyrolysis is often referred to as a gasification process where the gases evolved are the valued product. The solid remnant or slag resulting after the plasma treatment is relatively inert, has relatively high aggregate strength, and reasonably durable and thus can be used in construction applications.[65] The CO and hydrogen mixture resulting is called synthesis gas (syngas), which can be used for power production via combustion with roughly ½ the energy content of natural gas. Syngas can be used to make higher value products as well via addition of hydrogen to CO or CO_2 to make ethanol and methane. Furthermore, the CO formed can react with steam to produce additional hydrogen. Plasma pyrolysis however, being a thermal process is energy intensive and can require extensive upkeep. Additionally, the end products can be converted into fossil fuels so on some level, the process does not address the buildup of atmospheric CO_2. Plastics such as polypropylene produce gaseous propylene during pyrolysis and thus provides a means for depolymerizing and thus providing monomers for subsequent production of new polypropylene. Conversion efficiencies of up to 78% of polypropylene into gaseous products have been demonstrated with 94 percent of the product gas being propylene.[66]

Other plasma pyrolysis studies have shown solid polypropylene plastic to gaseous species in the form of predominantly hydrogen and acetylene by greater than 90 percent, showing the promise of plasma-based conversion of plastics wastes to useful feedstock.[67]

The disadvantages of thermal plasmas such as high energy expenditure and high capital and maintenance costs. The initial capital investment, power and maintenance costs must be less than the market value of the higher value products being produced. Getting to this point is the challenge for this promising technology.[68]

C. Opportunities for non-thermal atmospheric pressure plasmas

To date there has been only limited application of non-thermal atmospheric pressure plasmas for plastics degradation. Most of the work regarding NTAPs and plastics has been for surface modification in materials processing. Non-thermal plasma application to plastics recycling is relatively new and represents a frontier of possibilities owing to the control of plasma conditions and energies afforded by this plasma type. Indeed, it is possible to increase the recovery yield of monomers in conventional methods such as thermal pyrolysis via the introduction of a non-thermal plasma. These plasmas are associated with energetic electrons, excited species and radicals. These processes have the potential to break bonds to liberate monomer sections. In this case, plasmas can be used downstream of a conventional pyrolysis reactor to selectively drive the gas phase chemistry. This prospect was verified with HDPE, which constitutes some over 12% of all plastics waste. In a recent study a DBD discharge was used to process gas from the pyrolysis reactor, which resulted in a 55x improvement in the recovery of ethylene, a monomer of HDPE.[69] That work demonstrated the prospect of actively augmenting the gas phase process to improve yield. Such variations in gas phase chemistry is difficult with thermal reactors or even thermal plasmas for pyrolysis. It is possible to reform gaseous hydrocarbons derived from pyrolysis and convert them to hydrogen as well. Hydrogen, with its high energy density, is an energy carrier and has the potential for many applications such as electricity or heat production without the generation of global warming gases. Aminu and colleagues utilized a packed bed dielectric barrier discharge to use plasma in the presence of a catalysis to steam reform hydrocarbons.[70] In this case, a coaxial electrode geometry was used with ceramic beads containing transition metal catalyst particles such as nickel, cobalt, iron and copper located in the intervening gap. In such a discharge, shown schematically in Fig. 18, plasma is formed in the interstitial spaces of the catalytic particles. Surface ionization waves also travel along the surface of the particles. The field enhancements at the catalytic particle sites gives rise to enhanced plasma activity (ionization and localized heating) leading to enhanced chemistry in addition to

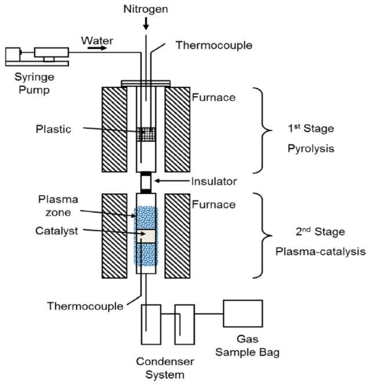


FIG. 18. Dual stage thermal pyrolysis-non-thermal plasma reactor for polymer treatment.[70]

surface chemistry that occurs at the surface of the particle.[71] In that work HDPE plastic was conventionally treated with a thermal pyrolysis reactor in the first stage to produce the pyrolysis gas. The gas then flowed into the DBD in the presence of injected water—to facilitate steam reforming for hydrogen production: $C_nH_m+nH_2O \rightarrow (n+m/2)H_2+nCO$ and $CO+H_2O \rightarrow H_2+CO_2$. The plasma processed gas was then captured in gas chromatograph sample bag. From the analysis of these samples, the plasma treatment stage gave rise to an enhancement in hydrogen yield showing potential for the method. The use of the non-thermal DBD plasma is desirable in that it allows for selectivity and overall reduction in energy consumption by adjusting voltage and waveform shape. The 2-stage approach taken here suggests a role for plasmas in augmenting conventional processing of plastics—and perhaps may evolve to first implementations of non-thermal plasma applications for plastics recycling. Further techno-economic analysis as well as additional optimization experiments are required to determine the relative performance of this two-stage method as compared to conventional pyrolysis.

Another example of non-thermal plasma augmenting conventional methods was demonstrated by Mancini and colleagues. Here the group was interested in improving the yield of conventional hydrolysis of PET plastic to precursor chemicals. In this case, the group demonstrated that exposure to PET samples to a low-pressure plasma improved the conversion rate by 40%! In that work, it was conjectured that surface modification by the plasma improved water uptake—making it more hydrophilic and thus more amenable to hydrolysis.[72] Direct conversion of pyrolysis products into higher quality oils (upscaling) has also been explored as described by Song and colleagues. In this work, pyrolysis products from polyethylene heated in a tube furnace served as feed gas for a cylindrical geometry, packed bed dielectric barrier discharge. The packed bed reactor consists of a central powered electrode with a coaxial grounded mesh. The dielectric barrier spanning the interelectrode gap in this case was a catalyst aggregate (SiO2/Al2O3 particles). Plasma is formed in the interstitial spaces. Nitrogen was the carrier gas.[73] Rorrer and colleagues demonstrated depolymerization of polypropylene using a packed bed dielectric barrier discharge as well. In this case, the packing dielectric was a mixture of catalyst particles and plastic particles in the presence of hydrogen to initiate hydrogenolysis. Hydrogenolysis is a chemical reaction in the presence of a catalyst that breaks carbon-carbon bonds accompanied by the addition of hydrogen to the molecule, leading to depolymerization. In this case the hydrogen plasma was key to realizing this reaction.[74]

The chief issue with plastic waste is its resistance to chemical erosion in the environment gives it an exceedingly long half-life. In general, plastics degrade very slowly in the natural environment by chemical, physical and biological means. Plastic biotic and biotic degradation processes that occur naturally in the environment are shown schematically in Fig. 19.[75]

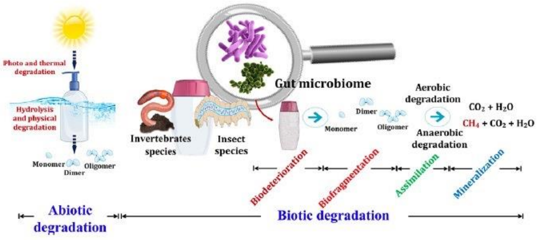


FIG. 19. Degradation of plastics in the environment via abiotic and biotic processes.[75]

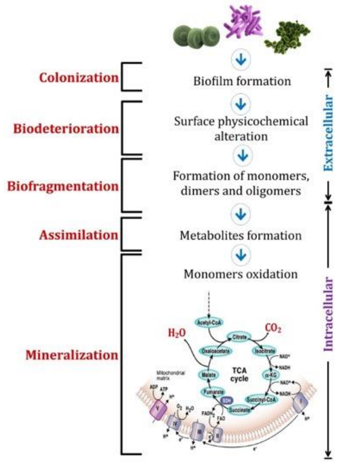


FIG. 20. Microbial degradation of plastics.[75]

Photo-driven degradation is considered the key process in abiotic degradation. Here light breaks bonds which lead to radical formation. These radicals then facilitate chain breaking leading ultimately to inert end species. In general, abiotic degradation provides an active substrate for biotic degradation processes, as microbes can metabolize the smaller molecular fragments formed abiotically. The biological pathway refers to degradation that is driven in part by the microbes that metabolize the plastic. Functionalization of the polymer surface can enhance the biological degradation rate. It has been shown that the rate of bacteria driven degradation of plastics depends on how adherent the cells are to the surface. Plastics are typically hydrophobic in nature. In general, oxidized hydrophilic surfaces are more amenable to bacteria proliferation.[76] Schematic depiction of plastic degradation and subsequent assimilation via microorganisms is shown in Fig. 20.[75] Note a key step is biofilm formation. Biofilm formation is followed by surface deterioration due to microbial metabolic activity, ultimately leading to assimilation and mineralization—reduction to carbon dioxide and water. Any process that reduces hydrophobicity favors biofilm formation and adherence to the surface followed by subsequent increased microbial activity and thus increased plastics degradation. Abiotic processes such as UV light exposure or external processes that affect functional group concentration associated with plastic surface chemistry can give rise to the transformation to a hydrophilic surface. Exposure to an air plasma can affect the surface energy of plastic material. Indeed, the plasma treatment of plastics to make surfaces more hydrophilic is well established for industrial application such as water repellent fabrics and surfaces. In this regard, plasmas, particularly non-thermal plasmas, can be used to pretreat plastics so as to enhance the natural rate of degradation of those plastics that are disposed of in a landfill. Figure 21 illustrates the effect of atmospheric pressure plasma treatment on polymeric powder (PET in this case). Here the powder is subsequently exposed to a liquid dye solution. Capillary action gives rise to a rise of the colored solution—with the more hydrophilic samples rising the highest over same time interval. Here it can be seen by this so-called Washburn Capillary Rise test[77] that the plasma treated sample on the left has become highly hydrophilic owing to plasma treatment in contrast to the untreated powder on the right. Scally and colleagues have been able to demonstrate that this surface enhancement favors biofilm formation is in fact valid.[78] They were able to show that LDPE when treated with a non-thermal, atmospheric pressure streamer discharge and subsequently immersed in a *Ps. aeruginosa* bacteria broth media, degraded at an accelerated rate. The multistreamer reactor used to treat the samples was operated in regular air as can be seen in Fig. 22. The group conjectured that the plasma pretreatment modified the surface making it more amenable to cell adhesion and subsequent proliferation.[78] In this regard, the role of plasma in this case is preparation for enhanced degradation thus playing a role in reducing plastics half-life and reducing overall the loading on landfills. This approach provides at least a near term solution to managing the proliferation and disposal

FIG. 21. In-house PET powder exposed to blue dye for Washburn capillary rise test, showing greatest rise in plasma treated simple on left suggesting increased hydrophilicity.[77] (R. Pinsky)

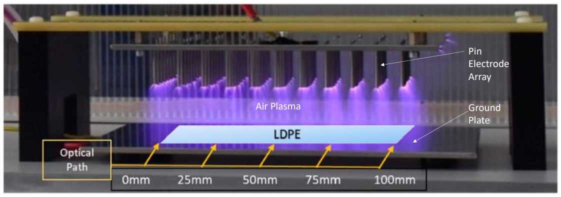


FIG. 22. Plastics plasma pretreatment reactor for surface functionalization in preparation for biofilm generation.[78]

of conventional plastics. In fact, it can be envisioned that this approach may be the next credible implementation of non-thermal plasmas for the treatment of plastics--surface modification for accelerated degradation and uptake in the environment.

Biopolymers are polymers produced by living cells in contrast to traditional plastics which is typically produced from petroleum. Examples of biopolymers include proteins (polypeptides) and starch (polysaccharides). Biopolymers naturally decay in the environment and thus represent a sustainable solution to the plastics problem. Biopolymers such as PLA (polylactide acid) is derived from essentially sugar is currently being used in the fabrication of a number of products that once featured petroleum derived plastics such as straws, lids and lids containers and cups. These products are biodegradable.[79] Depolymerization of naturally occurring biopolymers is also of interest in that the monomers alone have a range of applications particularly in the healthcare realm. This is of particular interest in that biomass for these molecules is large. Chitin for example is a biopolymer that makes up exoskeleton of insects and cell walls of fungi. This material can be used for edible packaging derived from biomass via depolymerization and subsequent polymerization to produce similar or higher value products from the environment.[80] Lignocellulose constitutes another class of abundant biopolymers. These polymers are derived from non-edible portions of plants and typically are disposed of in landfills. These too can be potentially depolymerized and then using the resulting monomer chains, fashioned into new polymers.[81] It was demonstrated that plasma generated in water could be used to depolymerize the biopolymer Alginate which is derived from brown algae. Authors of this study asserted that the reactive species produced by the plasma in water facilitated scission, giving rise to the release of monomers in solution. The interaction of plasmas with liquid water drives locally nonequilibrium conditions. Local temperature, species concentrations, photon deposition, kinetic energy of ions and electrons, pH, and ionic strength can all exist in states far from equilibrium.[45] Authors suggested that peroxide and ozone played key roles in the production of the monomers. This approach to depolymerization has the inherent advantage of being relatively nontoxic and potentially less expensive than conventional hydrolysis depolymerization where the polymers are treated in a high temperature acidic bath. In a conventional thermal bath, cleavage is stochastic with little hope of selectivity control.[82] As seen here, again plasma may play a role in the depolymerization of biomass feedstock for the purpose of generating new, biodegradable plastics. The fact that the experiments takes place in the liquid phase is desirable for extraction of monomers. The desirable aspect regarding the development of an economy based on biopolymers is that the products degrade, the feed stock--biomass is abundant, and do not rely on fossil fuels for production.

While not strictly a low temperature plasma but definitely a non-thermal, electron beams already play a key role in polymer processing as well. Electrons beams have widely been used in the polymer field for crosslinking, which involves bond breaking of one molecular chain to facilitate the formation of a bond between the break point and an adjacent monomer. This greatly improves the mechanical properties of polymers such as increased melting point and reduced deformation flow at high temperature. Indeed, this electron-based technology is now widely accepted by the wire and cable industry as a means to achieve these improved qualities without any thermal processing or chemicals. Polymers that cross link best are those that lack long chains such as those where the carbon is bonded to four neighbors (quaternary carbon). These include polymers such as polyethylene and PVC--common jacket material for applications such as wire insulation.[83] The electron beam can also drive scission leading to depolymerization for quaternary carbon polymers. It was recently reported that depolymerization of PET could be enhanced via electron beam irradiation. Jamdar and colleagues irradiated PET samples before conventional glycolysis based depolymerization.[84]

Here glycolysis refers to the degradation of PET by ethylene glycol in the presence of a catalyst. It should be noted that under conditions of very fast rise time, high voltage pulsed power-driven atmospheric pressure plasmas can produce energetic electron beams. In this regard, atmospheric pressure plasmas can potentially be used to driven e-beam related depolymerization as well.[85]

D. Future outlook

To address plastics, recycling is key. Chemical processes that takes in used, mixed plastics waste and then depolymerizes the aggregate to form precursors using electricity derived green energy is an aspirational recycling goal. Early experiments suggest that plasmas may indeed be able to play a role in such recycling. The plasma driven process is completely non-thermal with the bulk of the reactants and solvent solution residing at room temperature, so that energy loss is minimal. This is the potential promise of plasma-based processing of plastics. Taken altogether, plasma treatment of plastics holds some promise--an opportunity yet to be fully explored. Further investigation is certainly warranted, particularly in the area of non-thermal plasmas where selectivity and control are possible. Such control may allow for optimization of scission leading to high yield depolymerization. It is this very aspect that makes plasmas an interesting, transformative approach to the chemical recycling of plastics waste. Here the activation energy is provided by the plasma itself, produced by renewable electricity. The case for plasma treatment of plastics is certainly compelling. Its actual suitability from a scalability and cost standpoint requires sustained investment in studies that frame the role that plasmas can play.

IV. FOOD AND AGRICULTURE. THE CHALLENGE OF FEEDING SUSTAINABLY 10 BILLION PEOPLE.

With a world population of over 7.8 billion, which will reach 9.7 billion in the next 30 years,[22] the global demand for food increases steadily thus notably raising the pressure on the natural resources and environment. In 2050, food demand is expected to increase by more than 50 percent.[86] As stated by the Food and Agriculture Organization (FAO) of the United Nations,[87] in addition to this challenge, food and agriculture production systems worldwide are facing other unprecedented concerns related to "*rising hunger and malnutrition, adverse climate change effects, overexploitation of natural resources, loss of biodiversity, and food loss and waste*".

Therefore, expanding and accelerating the transition towards sustainable food and agriculture is already urgent beyond any doubt. The FAO states that sustainable agriculture "*must nurture healthy ecosystems and support the sustainable management of land, water, and natural resources while ensuring world food security*".[88]

Feeding nearly 10 billion people in a sustainable way by 2050 is a major challenge, which has been worked on over the last decade. Recently, Vågsholm and colleagues have published a comprehensive study on this topic, revealing the need to make the right trade-offs between sustainability, food safety, food security, and making better use of already produced food, if we are to be able to respond to food demand in 2050 in a sustainable way.[89] Here, food safety refers to those conditions and practices while handling, preparing, and storing of food, that preserve its quality in terms of preventing contamination and foodborne illnesses. On the other hand, food security was defined in the Rome Declaration on World Food Security (1996 World Food Summit) in the following terms: "food security exists when all people, at all times, have physical and economic access to sufficient, safe and nutritious food to meet their dietary needs and food preferences for an active and healthy life".[90] Food security relies on four pillars, namely: (a) availability of food (sufficient food production), (b) access to food (ability of individuals and households to secure sufficient food), (c) utilization of food (ability of individuals and households to maintain a healthy diet that meets human requirements in terms of nutrition), and (d) the stability of these three mentioned conditions.[91] Food security is a major and persistent concern in many developing countries, where the supply of nutritious and safe food is usually limited, and the purchasing power of consumers is low. The challenges to food security are mainly related to poverty, although there are some others that include the increasing diversion of edible crops towards biofuel production (which contributes to the reduction of food availability) and the rapid urbanization that makes more and more consumers depend on the purchase of food.[91]

A. The "menu of solutions"

Vågsholm et al propose that meeting the challenge sustainable food for everybody in 2050 will require: (1) changes of human food chains in the next 20 years (e.g. shifting human food demand to more plant-based diets could help), (2) reduction of food loss and waste, (3) intensification of food production, (4) redistribution and reprocessing of food, and (5) recycling/recovering of lost and wasted food.[89] The EAT Lancet Commission on healthy diets from sustainable food production has also analyzed this challenge,[92] concluding that it will be possible to feed a global population of nearly 10 billion people a healthy diet within food production boundaries by 2050 provided there is a substantial global shift towards healthy dietary patterns, large reductions in food loss and waste, and major improvements in food production practices. They suggest several dietary changes and recommend doubling the consumption of fruits, vegetables, nuts, and legumes and halving the consumption of red meat and sugar. Thus, they put the focus on plant-based foods.

On the other hand, the World Resources Institute has elaborated a detailed report, the World Resources Report (WRR) addressing the challenge of creating a sustainable food future heading into 2050.[86] WRR presents an in-depth analysis detailing three specific challenges facing the global food system: (i) food supply, (ii) land use and (iii) greenhouse emissions. This report proposes a 22-item “menu of solutions” that could deliver a sustainable food future. These solutions are classified into five categories aiming at: (1) reducing growth in demand for food and other agricultural products, (2) increasing food production without expanding agricultural land, (3) protecting and restore natural ecosystems and limit agricultural land-shifting, (4) increasing fish supply, and (5) reducing greenhouse gas emissions from agricultural production. Among these twenty-two categorized potential solutions, several themes stand out including to raise productivity, slow demand growth, link agricultural intensification with natural ecosystems protection, moderate ruminant meat consumption, target reforestation and peatland restoration, reduce Greenhouse Gases (GHG) emissions from agricultural production sources, and spur technological innovations.

B. Opportunities of non-thermal-plasmas in supporting future food security

Innovation is considered as key for achieving food demands in the near future in the World Resources Report.[86] Non-thermal plasma technology could be a useful tool helping to undertake some of these WRR solutions in a sustainable way. Due to the great extension of the subject, we will mainly focus on agriculture, as one part of the solution will be consuming plant-based foods. One of the first connections of plasmas with agriculture was published as a patent by Krapivina and colleagues in 1994,[93] who reported for the first time on the modification of seed germination performance using a cold plasma reactor. Since then, exciting research on different topics of what was termed as *Plasma Agriculture* have been carried out and compelling results have aroused from them.[94]

In addition to Food safety (Subsection B.1.), which is a cross-cutting issue, we have selected only those items in the WWR for which plasma technology could play a role and could be, at least, part of the solution, including Reducing food loss and waste (Subsection B.2.), Increasing food Production without Expanding Agricultural Land (Subsection B.3.), and Managing effluents and wastes from agricultural and food activities (Subsection B.4.). Finally, in Subsection B.5. we will describe the use of *plasma activated water* as a possible way to scale-up plasma treatments.

B.1.Food safety

A major challenge has emerged in recent years related to food safety. Controlling new and existing infectious diseases is one of the utmost public health challenges facing nations worldwide. Foodborne illness continues to be a leading cause of morbidity and mortality, generating substantial socioeconomic losses globally.[95] Food contamination problems produced in developing countries are even greater, due to lack of access to clean water for food preparation, poor transportation, inadequate storage infrastructure, poor personal hygiene, and improper handling.

Fruits and vegetables are essential in a healthy human diet. Nevertheless, they can be carriers of foodborne pathogens, which can affect their quality as well as cause serious health problems.[96-98] In the last years, raw or minimally processed fresh produce consumption has notably grown and so has the number of foodborne illness outbreaks.[99] Centers for Disease Control and Prevention (CDC), in its 2017 Annual Report, reported that 841 foodborne disease outbreaks occurred in the United States that year, resulting in 14,481 illnesses, 827 hospitalizations, 20 deaths, and 14 food product recalls. *Norovirus* was the most common cause of confirmed, single-etiology outbreaks, accounting for 140 (35%) outbreaks and 4,092 (46%) illnesses. The next most common cause was *Salmonella*, accounting for 113 (29%) outbreaks and 3,007 (34%) illnesses, followed by Shiga toxin-producing *Escherichia coli* [causing 19 (5%) outbreaks and 513 (6%) illnesses], and *Clostridium perfringens* [causing 19 (5%) outbreaks and 478 (5%) illnesses].

Noroviruses are highly infectious human pathogens and a major cause of diarrheal illness worldwide.[100] Norovirus infections can be asymptomatic, but also could lead to severe, life-threatening gastroenteritis. According to the Center for Disease Control and Prevention (CDC) of the U.S. Department of Health & Human Services, each year, on average in the United States, norovirus causes 19 to 21 million cases of vomiting and diarrhea illnesses and 2,270,000 outpatient clinic visits annually.[101] 40% of norovirus infections are estimated to be caused by the consumption of contaminated foodstuffs.[102] Norovirus is responsible for most of the foodborne outbreaks associated with fresh fruits and vegetables in the United States and European Union.[103] All this gives us an idea of the dimension of this problem.

In 2020, the FDA investigated in the USA multistate outbreaks of *E. coli* O157:H7 infections linked to leafy greens, and *E. coli* O103 infections linked to clover sprouts. It also reported about multistate outbreaks of *Salmonella* Stanley infections linked to wood ear mushrooms, of *Salmonella* Enteritidis infections linked to peaches, of *Salmonella* Newport infections linked to red onions, of *Cyclospora* infections linked to salad products (made by a fresh food company containing iceberg lettuce, red cabbage, and carrots), and of *Listeria monocytogenes* infections linked to enoki mushrooms.[104] In 2019 (without the problem of the COVID-19 pandemic yet) the number of multistate outbreaks investigated by the FDA in the USA was even higher.

On the other hand, the food industry also faces the difficult challenge of producing mycotoxin-free food due to the inefficiency of traditional mycotoxin removal strategies. Mycotoxins are low weight molecules produced as secondary metabolites by certain fungi and they induce a toxic response when introduced at a low concentration to higher vertebrates and other animals via natural route.[105] Mycotoxin-producing fungi can invade the food at any stage of the food production process (before harvesting, harvesting, drying, and storage), exposing consumers to the risk of contamination. The effects of some foodborne mycotoxins can appear rapidly and acutely, however, rather than outbreaks, mycotoxin contaminated food are usually related to longer-term chronic or cumulative health issues, including the induction of cancers, immunodeficiency or teratogen effects.[105,106] Fungi such as Aspergillus, Fusarium, Penicillium, Claviceps, and Alternaria spp. colonize their host and produce mycotoxins. Among the wide variety of mycotoxins identified, thirty are considered a threat to human or animal health. Mycotoxins cannot be detected with the naked eye, and they do not have an odor or modify the organoleptic properties of the food, which makes them difficult for the consumer to detect. Agriopoulou and colleagues recently published a work reviewing the occurrence and importance of mycotoxin in foods.[107] Prevention and detoxification are two main strategies used to control mycotoxin contamination of foods. The use of fungicides for control of fungal infection, the use of antagonistic fungi, the control of environmental factors during seeds storage (temperature, moisture level, and humidity of warehouses) avoiding mold growth and mycotoxin production, are pre-harvest strategies to prevent mycotoxin contamination pre-harvest. Post-harvest strategies for decontamination of mycotoxins from agricultural products include biological, chemical, and physical methods. The use of conventional processing technologies such as cooking, boiling, baking, frying, baking, and pasteurization, most mycotoxins remain chemically and thermally stable, so that for complete mycotoxin elimination, other strategies must be used.[107] It has been shown that radiation partially removes mycotoxins in foods, as at high dose it affects the quality of commodities. Control through the use of chemicals such as ammonia, hydrated oxides or chitosan have been successful for mycotoxins decontamination. The main disadvantage here is usually these chemicals can cause secondary contamination and have harmful effects on the nutritional value of the products. Ozonation is another technology used to detoxify mycotoxins. This is a simple and very efficient approach, which does not leave harmful residues after application. On the other hand, research conducted in the last 20 years has shown that detoxification/degradation of mycotoxins by biological processes is a promising alternative approach as it can lead to the production of less or even zero toxic end products and intermediates.[107]

Plasma-based decontamination of foods

Thus, there is an urgent need to develop additional green techniques to disinfect foodborne pathogens on the surface of fruits and vegetables to ensure food safety. In this scenario, non-thermal plasma technology emerges as a promising approach to fresh agricultural produce decontamination due to its ability to generate an abundance of active species with antimicrobial attributes at ambient conditions.[108-110] Several types non-thermal atmospheric pressure plasmas (frequently named Cold Atmospheric Plasmas (CAPs), as their temperatures are close to the room temperature) have been developed for this purpose. NTAP decontamination mechanisms are mainly ascribed to highly reactive oxygen and nitrogen species (ROS and RNS) created within the plasma (OH•, H•, O•, NO, H_2O_2, NO_x, O_3, NO_2^-, NO3−, peroxynitrite…), as well as UV radiation, all of them inducing highly oxidizing effects. NTAP treatment can be performed both directly[99,111-114] and indirectly using the so-called plasma activated water (PAW),[115] which contains ROS and RNS formed upon plasma treatment.

There are already numerous studies carried out in this regard most of them gathered in Varilla et al review.[116] Just to cite a few examples, Critzer et al reported on the efficiency of using a one atmosphere uniform glow discharge plasma for inactivation of Escherichia coli O157:H7, Salmonella, and Listeria monocytogenes on apples, cantaloupe, and lettuce, respectively.[117] El Shaer and colleagues used a gliding arc for decontamination of Erwinia *carotovora* subsp. *carotovora* strains spread on potato slices, showing that this was an effective and quick approach.[118] New South Wales Department of Primary Industries laboratory reported in 2018 that a new plasma device developed to try to eliminate microbial contaminants such as Salmonella, Listeria, and E.coli, was being trialed.[119] Han et al evaluated the efficiency of a surface dielectric barrier discharge in the inactivation of three foodborne pathogens, E. coli O157:H7, S. Typhimurium, and L. monocytogenes, in fresh vegetables, fruits, nuts, and powdered food samples,[99] showing that surface roughness is an important factor for the antimicrobial efficacy of cold plasma treatment. Zhao and colleagues have also designed a DBD based plasma reactor allowing surface decontamination (Staphylococcus aureus) of rolling fruits (see Fig.23).[96]

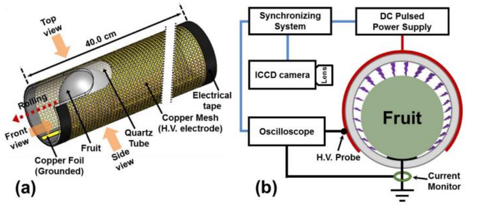


FIG. 23. Atmospheric pressure air dielectric barrier discharge for decontamination of rolling fruits.[96]

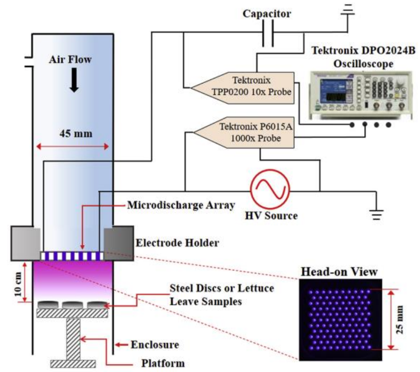


FIG. 24. Air plasma generated by a two-dimensional array of integrated coaxial-microhollow dielectric barrier discharge, designed for food decontamination.[121]

Cold plasmas have been also shown to be a non-thermal food processing technology to decontaminate viral pathogens (including Norovirus) in meat and fresh vegetables.[110,120,121] In Bae et al. work,[120] the use of an atmospheric pressure plasma was investigated against murine norovirus (MNV-1), as a norovirus (NoV) surrogate and hepatitis A virus (HAV) associated with three types of fresh meats (beef loin, pork shoulder and chicken breast), and it was shown that a 5 min long treatment results in > 99% reduction of MNV-1 titer and > 90% reduction of HAV titer without concomitant changes in meat quality. Aboubakr and colleagues found a > 5 log10 reduction in the titer of feline calicivirus (FCV) on Romaine lettuce leaves after 3 min wet exposure to air plasma generated by a two-dimensional array of integrated coaxial-microhollow dielectric barrier discharges (see Fig. 24).[121]

Recently, cold plasma technology has been also proposed as an effective post-harvest technique for the degradation of mycotoxins and inactivation of mycotoxin-producing fungi.[122,123] L. ten Bosch et al. have shown the efficacy of a Dielectric Barrier Discharge operated with air for the degradation of selected mycotoxins (deoxynivalenol, zearalenone, enniatin, fumonisin B1, and T2 toxin produced by *Fusarium spp.*, sterigmatocystin produced by *Aspergillus spp*. and AAL toxin produced by *Alternaria alternata*).[124] They suggest toxin degradation results from a combination of different mechanisms such as chemical reactions with reactive species generated in the plasma volume such as O, O_3, OH, and NO_x and/or decomposition after collision with electrons and ions leading to cleavage of molecular bonds. A careful analysis of the main advantages of using cold atmospheric plasma technology versus classic methods in mycotoxin decontamination has been performed by Hojnik and colleagues, which have emphasized that CAP mycotoxin decontamination of food not only overcomes many of the disadvantages and obstacles of physical, chemical and microbial decontamination procedures, but they also offer a more economically favorable and environmentally benign solution, with a negligible effect on the quality of many types of treated food (as plasma species enable a high decontamination efficiency in a very short time compared to alternative decontamination methods).[125]

On the other hand, in the last decade, non-thermal plasma technology has emerged as a powerful tool for surface decontamination not only of foodstuffs but also of food packaging materials.[126] Non-properly sterilized food packaging may cause contamination of the food and consequently lead to health risks and economic losses. Conventional sterilization methods (including dry heat, steam, UV light and chemicals), traditionally used for packaging materials in the food industry, have as a main drawback the generation of liquid effluents whose management increases the cost of the process. Non-thermal plasma sterilization is a chemical free, fast, and safe approach, applicable to a wide range of packaging materials and does not result in any residues. However, its adoption in the food packaging sector is limited by the treatment times, which often extend to minutes.

B.2. Reducing food loss and waste

According to WRR, nearly 24 percent of all food produced (measured by caloric content) is lost or wasted from farm to fork.[86] About 56 percent of food loss and waste happens in developed regions and 44 percent in developing regions. Food loss and waste not only results in losses of almost $1 trillion per year, but it also compromises food security (especially in developing countries) and wastes natural resources (by consuming about one-quarter of all water used by agriculture each year, requiring an area of agricultural land greater than the size of China, and generating about 8 percent of global GHG emissions annually).[86]

Vagsholm and colleagues claim that more than one billion additional persons could be fed by eliminating global food waste and loss, and also argue the reduction of food lost or wasted would lead to more efficient land use and better water resource management with positive impacts on climate change, livelihoods, and sustainability.[89] The loss and waste of food leads to the loss of work, money, water, energy, land and other resources that were used to produce the food, thus compromising sustainability.

Food loss and waste occur at the different stages of the food supply chain, that is, during: (1) production or harvest, (2) handling and storage (because of degradation by pests, fungus, and disease), (3) processing and packaging, (4) distribution and marketing (edible foodstuffs discarded because it is noncompliant with aesthetic quality standards or is not sold before "best before" and "use-by" dates), and (5) consumption (food purchased but not eaten).

In developing countries, large storage losses occur due to limited refrigeration and food processing. To reduce pest damages during storage, improved the long-term resistance of products to spoilage, and the control of crop diseases are some relevant aspects to consider in order to reduce food loss and waste.

Losses incurred by insect infestation during storage of agricultural products must also be considered at the post-harvest stage. For example, *Plodia interpunctella* (Indian meal moth) is one important pest worldwide thriving in tropical habitats. It is frequently found at grain storages and its infestations cause both direct commodity loss and indirect economic costs through pest control costs and quality losses.[127] *Tribolium castaneum* is another worldwide insect pest of stored products, and the worst of flour mills .[128,129] It feeds on cereals, flour, starchy material, fruit nuts, millets and prepared cereal foods, being one of the most economically important insect pests of processed food.

Ethylene gas is a natural plant hormone that regulates a large number of physiological processes in plants, which can also induce negative effects on fruits and vegetables such as accelerated quality loss, over-ripening, increased fruit pathogen susceptibility, and senescence, among others, with a reduction of freshness and shelf life. Ethylene gas is released by fruits and vegetables during postharvest storage.[130] There are also external sources of ethylene occurring along the food chain, in packages, storage chambers, during transportation, and domestic refrigerators. To remove ethylene surrounding fruits and vegetables is a major challenge in the postharvest chain in order to avoid detrimental effects on the quality of these commodities and keeping them fresh. Technologies used to control ethylene include: physical methods (adsorption and absorption), chemical methods (such as catalytic decomposition, thermal incineration, O_3, NO, and ClO_2 treatments), and biological methods (aimed at blocking various enzymes and precursors involved in ethylene synthesis), all having their downsides for removing ethylene from agricultural storage facilities.[131]

Finally, the adequate management of diseases and infestations of crops is another important challenge that needs to be urgently tackled in order to reduce food loss and waste, which would help to ensure food security in developing countries. It is estimated that between 20 and 30% of crops are lost due to plant diseases.[132] Seeds contaminated with pathogenic fungi and bacteria are the main source of many plant diseases worldwide,[133] causing significant food and economic losses each year.[134] Moreover, pathogen-harboring seeds are an important pathway for the spread of plant diseases both locally and over long distances. There is a risk of plant pathogens transmission to new locations in interstate and international seed trade, which is a particularly sensitive issue when it comes to developing countries with limited resources. Seeds are also a vehicle for the introduction of pathogens into the soil.[135] Thus, the use of pathogen-free seeds constitutes a preventive disease control strategy. On the other hand, foliage and root diseases are serious plant diseases in agriculture due to difficulties in controlling them. The infection of leaves and roots with bacteria and fungi can cause a significant reduction in the quality and yield of the crops.[132]

Plasmas for pest control, preservation and freshness-keeping

Research done so far shows that cold plasma could be a useful technique for stored product insects' control in the near future. Non-thermal plasmas have been shown to have insecticidal activity against larval and pupal stages of *Plodia interpunctella* and *Oryzaephlius surinamensis*.[127,136] This technology has also demonstrated to be an efficient tool for the control of *Tribolium castaneum*.[129]

On the other hand, due to the microbicidal action of plasmas containing oxygen/air, they can also be used for shelf-life extension of heat-sensitive fresh food products.[137] In this way, cold plasma technology has been often proven in the inactivation of fungal pathogens contaminating harvested vegetables and fruits.[132] Contamination with *A. flavus* and *A. parasiticus* of hazelnuts, pistachio nuts, peanuts, and maize can be reduced using cold plasma treatment. Longer preservation of grapes, bananas, lemons, blueberries, date palms, and citrus fruits has been reported upon plasma treatments.[132] Non-thermal plasma treatments have shown to be highly efficient for decontamination of fresh products at ambient temperature (including meet, fruits and vegetables) even inside sealed packages.[114,138,139] Figure 25 depicts a schematic of a DBD reactor used for in-package treatment of foods.[137] Here ozone generated with the plasma assistance is expected to be one of the key factors contributing to antimicrobial efficacy of plasma approach. High efficacy and lability of ozone makes these *in-package* treatments a promising and excellent strategy (as it does not require unpacking) that would help to increase shelf-life of food commodities, so aiding to reduce food loss and waste. But non-thermal plasma technology would not be only useful to treat food directly, it can be an excellent prophylaxis strategy used to clean the air of agricultural storage facilities due to its microbicidal activity.

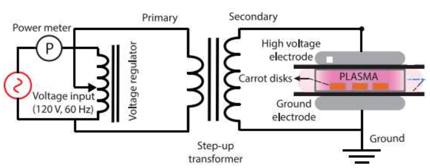


FIG. 25. Experimental set-up for in-package DBD plasma treatment of carrot disks.[137]

Active food packaging also contributes to the reduction of food losses. The main active packaging techniques involve the application of substances that absorb oxygen, ethylene, moisture, carbon dioxide, flavors, odors and those that release carbon dioxide, antimicrobial agents, antioxidants and flavors.[140] The immobilization of bioactive functional compounds on food packaging such as probiotics, proteins, and vitamins, helps to improve the properties of packaged foods (mainly fresh fruits and vegetables). NTAP technology makes it possible to immobilize such bio-active coatings.[126] Also, antimicrobial substances like chitosan, silver and triclosan have been immobilized on films by plasma treatment. The use of these bio-active/antimicrobial films extends the shelf life of foods, helping to address food loss and waste issues. On the other hand, NTAP treatment of food packaging can be used to modify their barrier properties in order to control mass transfer through the package, which is also a crucial factor in the shelf life of food. At present, not only are there published some works related to the plasma-assisted deposition of thin films on PET foils in order to reduce the passage of gases,[126] but also plasma technology for this use is even commercialized.[141]

In 1998, Graham and colleagues demonstrated for the first time the capacity of plasmas for removal of ethylene in fruit and vegetable storages.[142] NTAPs are capable of easily and quickly breaking down ethylene at ambient pressure and low temperatures (advisable to preserve food quality), but because of its poor selectivity for carbon dioxide it could result in unwanted byproducts (aldehydes and organic acids). This is why, more recently, the use of NTAP reactors in combination with catalytic/photocatalytic films has been proposed, showing a high efficiency for removing ethylene from agricultural warehouses.[132,143,144]

In summary, non-thermal plasmas are a very promising non-thermal processing technology to decontaminate and preserve fruits and vegetables. The effectiveness of plasma treatment depends on the rugosity of their surfaces,[110] but this is an aspect still under research. Also, further investigations on the possible impact of plasma treatment on organoleptic and nutritive qualities of these commodities are needed, as well as, to improve still little knowledge available about potential cytotoxic and mutagenic effects associated with plasma treatment,[145] as limited research has been conducted on the safety of plasma-treated fruits and vegetables for human or animal consumption.

Plasmas for crop disease control

The decontamination of seeds while in storage before planting notably improves the survivability of planted seeds and to promote their healthy growth. Conventional chemical seed treatment methods for reducing pathogen contamination as fungicides and bactericides offer a narrow range of effectiveness in controlling only certain pathogens. Other important disadvantages of these methods are the development of resistance to fungicides and bactericides, the risk the chemicals used for seed treatments pose to animal and human health, and difficulties to germinate of seeds whose surface is coated by chemicals (worsening planting success). In this scenario, plasma-assisted treatment arises as a compelling alternative to decontaminate seeds, as it is effective against a wide range of pathogens, nontoxic to humans and animals, nonpolluting, and easy to apply. It allows decontamination of pathogenic fungi, bacteria, biofilm formers, viruses, and insect pests, and has much less possibility to induce resistance from pathogens.[132] Many species generated by plasma are unstable species, so the impact of plasma on the environment is short-lived. A detailed review on the use of cold plasma technology in the control of plant diseases has been recently published.[132]

Wheat, rice, and maize are the three most important food crops in the world. Fungi species in the genera of *Aspergillus*, *Penicillium*, and *Fusarium* are among the most predominant pathogens contaminating these seeds during storage,[146] which is a major issue to be addressed as they not only are toxin-producing fungi with effects on human and animal health effects, but also because they do affect the viability and yield of the crop. Plasma treatment has been demonstrated to be effective for the decontamination of *Aspergillus* and *Penicillium* species in seeds of tomato, wheat, bean, chickpea, soybean, barley, oat, rye, lentil, and corn. It is also effective against *Gibberella fujikuroi* (*Fusarium*) fungi causing bakanae disease in rice seeds,[147] and *Fusarium culmorum* in wheat and barley seeds.[148,149]

Seed damage could be also caused or enhanced by storage insect pests, which usually results in seed germination reduction.[150] Insect infestation is an issue of high importance affecting seeds (at both pre-harvest and post-harvest stage) and agricultural products in general, as already described. Plasma technology has demonstrated to be also useful in this respect. M. F. Abd El-Aziz and colleagues have demonstrated the efficiency of a pulsed atmospheric-pressure plasma jet for control of the Indian meal moth *Plodia interpunctella*.[127]

FIG. 26. Atmospheric plasma jet in direct contact with a plant leaf infected with pathogens.[151]

Compared to seeds, leaves and roots are relatively more difficult to treat with plasma, thus results from this type of plasma treatment on infected leaves or roots has been much less reported.[132] Zhang et al reported on the ability of an atmospheric cold plasma jet to cure fungus-infected plant leaves and control the spread of infection (see Fig. 26), concluding it is an attractive tool for plant disease management.[151] Plasma activated water could be also used with this purpose because of its efficacy addressing these plant infections. Moreover, it offers an easy to scale-up approach when it comes to controlling crop diseases. Very recent studies show that PAW treatment enhances the plant defense responses and provide an encouraging framework for future applications in plant disease management programs.[152]

Finally, air quality is an important factor affecting the success of greenhouse crops. Because it is a very effective method for air purification, plasma technology is currently used to clean air in greenhouse facilities, removing impurities (including volatile organic compounds) from the air in addition to reducing mold growth and bacteria, and even odors. Omniaire 600N HEPA Air

Purifier, Novaerus Defend 1050 (NV1050), and Plasma Air 51E / 52E are some of devices already available in the market.[153,154]

B.3. Increasing Food Production without Expanding Agricultural Land

The expansion of the area of agricultural land causes changes in its location, varying from one region to another (e.g., from temperate areas to the tropics), and these changes in land use increase GHG emissions and cause loss of biodiversity.[86] Therefore, additional food must be produced through yield growth (intensification) not through the expansion of agricultural land, thus protecting natural ecosystems like forests and savannas. Agriculture has long been considered as the main cause of biodiversity loss and is likely that it continues to be so in the future if no major changes occur. Increasing the efficiency of the use of natural resources is the most important step to meet food production and environmental goals.[86] Among others, this means increasing crop yields over historical (linear) rates.

In the last sixty years, the use of fertilizers and scientifically bred seeds and the increase of the irrigation areas, have driven the continuous progress of crop yield. According to the World Resources Report, both crop output per hectare and milk and meat output from ruminants per hectare must grow each year more than they did historically if we are to avoid net land-use expansion.[86] Although nowadays yields are determined by a mix of different inputs including fertilizers, water, seeds, machinery, labor, and land, this report considers advancing in crop breeding as an important strategy to boost yields.

Plasmas to boost crop yield

The treatment of seeds using plasma technology allows surface modifications intended for increasing plant growth and/or alleviating the adverse effects of environmental stressors such as drought, waterlogging, high salinity, extreme temperatures, or pathogen infections, on germination.[155] Etching, introduction of functional groups, deposition of coatings, cleaning, and sterilization are typical plasma-assisted seed surface treatments. Some of them can change the wetting properties of the seeds, affecting the water uptake, which could be key when sowing. In this way, plasma treatment can make the seed surface (usually hydrophobic) hydrophilic, which favors the water uptake.[92] Also, delayed germination through retardation of imbibition helps to prevent chilling damage of seeds sown in cold and wet soils, which can be achieved through hydrophobic coatings on seeds.[156] Fluorocarbon containing plasmas (e.g. octadecafluorodecalin) are used for hydrophobic film deposition. In contrast, increasing the hydrophilicity or permeability of the seed coat (through hydrophilic coatings or etching, respectively) can give good results when sowing in water-scarce soils. As the thickness and permeability of the coat affects seed germination, seed coats partially etched by plasma action increase the water uptake thus favoring germination.[157] These treatments also promote accelerated germination, which could faster the crop emergence and increase the seedling development and are especially useful for slow-to-germinate seeds. In another context, the treatment of seeds with nitrogen-containing plasmas (e.g. aniline) also boosts accelerated germination together with early plant growth, as they could improve early nutrient (nitrogen, in this case) uptake.[156] This acceleration helps to the success of the crop when planting under adverse environmental conditions.

Non-thermal plasmas can be also used to remove toxic chemicals from the surface of the seeds (primarily used for the treatment of microbial contamination and germination enhancement, but degraded after long periods of storage) rendering them unsuitable for planting.[155] Unlike wet chemical removal of fungicides and insecticides, this is a sustainable method not requiring large amounts of water and organic solvents, or expensive drying technologies. Microbial contamination of seed surface could also affect seed germination and growth, so microbicidal action of plasma should be also helping. On the other hand, changes in seed physiology and at a molecular level caused by plasma treatment have been reported, and it is these that are currently garnering the most attention in this field of research.[155] Indeed, active particles from plasmas enter across the seed coats and directly interact with the cells inside, which might result in the stimulation of certain natural signals, hormones, and enzyme activities, thus enhancing germination and seedling growth.[158] Gómez-Ramírez et al. have reported an astonishing increase of Quinoa seeds germination from 60 % till nearly 100 % upon plasma treatment.[159] Figure 27 schematizes they used for seed treatment.

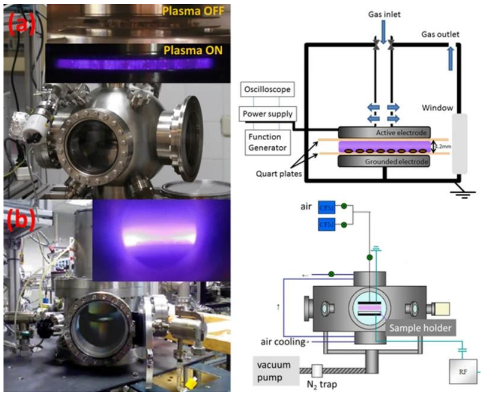


FIG. 27. DBD (a) and RF (b) reactors (and their schematic) for seeds treatment.[159]

But even though the mechanisms underlying these effects of cold plasma technology on plant germination and growth have not yet been fully deciphered and are still under study, the results found so far on improved germination, vigor, and health, without leaving chemical residues, support the idea that the plasma approach is a promising sustainable technology that will help to boost agricultural productivity.

Soil remediation by plasmas

The contamination of soil not only strongly influences its productivity, but could also affect air and water quality, being a more general environmental issue worldwide, not only limited to agriculture as explained by the Joint Center Research of the European Soil Data Centre.[160] It is worth it to point that 33 % of the land is highly or moderately degraded, as reported in Zhang et al work,[161] which gives an idea of the magnitude of the problem. Erosion, acidification, hardening, salinization, and chemical contamination are the main causes for it. In China, the Report on the National General Survey of Soil Contamination published by the Ministry of Environmental Protection and the Ministry of Land and Resources in 2014 showed that 19.4%

of soil survey points in agricultural land exceeded Level II requirements of the Soil Environmental Quality Standard (GB 15618-1995).[162] Major pollutants found were cadmium, mercury, arsenic, copper, lead, nickel, dichlorodiphenyltrichloroethane (DDT), and polycyclic aromatic hydrocarbons (PAHs). Organic pollutants such as organochlorine pesticides, polychlorinated biphenyls, phthalate esters, PAHs, and total petroleum hydrocarbons are often present in soils.[163] They are characterized by high toxicity, persistence, and bioaccumulation in the environment.

Although this is still a very recent field of study and there is much work ahead, so far, plasma technology has demonstrated noteworthy potential for soil remediation. In this way, the decontamination of heavily petroleum-polluted soils and organic-polluted soils has been successfully achieved.[164-166] Zhang et al have carefully reviewed the use of non-thermal plasma technology for organic contaminated soil remediation,[161] being DBD, pulsed corona plasma and gliding arc fluidized bed mainly used for this purpose. Compared to conventional technologies, non-thermal plasmas have low-energy cost, do not need the addition of further chemicals, can address remediation of most types of organic compounds, and is a fast method, being the scale-up of this technology the main drawback that still needs to be solved. Figure 28 is a schematic of a plasma reactor used for soil decontamination.

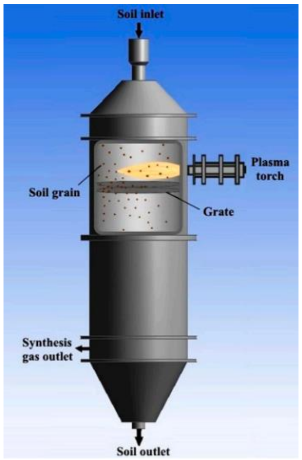


FIG. 28. Plasma reactor for soil decontamination.[166]

NTAP technology is also useful for microbial contamination of soils. Crop contamination by soil-borne pathogenic microorganisms is a severe problem resulting from fertilizing the soil with organic matter or the use of treated wastewater, often leading to serious infection outbreaks.[167] Therefore, disinfection of agricultural lands is also required for plant protection and improvement of crop quality and yields. Chemical and physical disinfection methods are currently used. Chemical methods are based on the direct addition of toxic chemical reagents (such as chloroform, ethylene oxide, bromomethane, mercury chloride), which are absorbed by crops and also leave residues that can contaminate the soil and water sources. Some of these products are even carcinogenic. Physical methods such as autoclaving and gamma irradiation consume large amounts of energy, and soil solarization based on the use of polyethylene sheets covering the soil to capture heat during summer months requires long times of application (about 2 months).[167] Lazra and colleagues have proposed the use of an atmospheric pressure plasma corona reactor for soil disinfection.[167] Even though this is a very new topic under research and improvements are needed (including the scale-up), so far these authors have shown promising results on reducing soil bacteria.

B.4. Managing effluents and wastes from agricultural and food activities

Proper management treatment of effluents and wastes from agricultural and food activities is another major challenge that needs to be sustainably addressed. The efficient use of water and its safe release to the environment is a key issue as already outlined in this paper. Degradation of pesticides, herbicides, and other micropollutants from agricultural water would be an optimal solution for its remediation, being this other important field where plasma technology can make an outstanding contribution. Since this topic has already been covered in depth previously in Section II, we just only mention that Bilea and colleagues have recently studied the potential of plasma treatment as a water reclamation process for irrigation and proved it is a good candidate.[168]

Another important issue for the agricultural and food sectors to face in this century is the appropriate management of food waste.[169] In the agricultural production process, there are losses due to mechanical damage and/or spillage during harvest operation (e.g. threshing or fruit picking), crops sorted out post-harvest…[170] Also, in post-harvest handling and storage, there are additional losses as those due to spillage and degradation during handling and storage. Food wastes (including those of vegetable origin) are rich in a wide variety of organic constituents (starches, oils, proteins, fats, phosphates, nutrients, amino acids, and natural acids) and the valorization of these wastes to different sorts of biofuels (biodiesel, bioethanol, biohydrogen, bio-oil, biochar, and biomethane) through efficient technologies can be an attractive and viable approach contributing to reduce the dependency of the energy sector on fossil fuels, and one of the most promising ways to contribute to sustainable development,[171] as it also helps to reduce the environmental burden of food waste disposal, lessen depletion of natural resources, minimize risk to human health and maintain an overall balance in the ecosystem.[172] Plasma technology can be also useful for food-waste-to-energy conversion, but for this purpose more energetic plasmas are needed. Thus, thermal plasmas have been successfully utilized for the gasification of carbon-based wastes so they can be also considered an appropriate and promising approach to food waste addressing, although this is still an unexplored area of plasma applications.[173]

On the other hand, these more energetic plasmas would also be useful to treat manure produced in confined animal facilities, which is one of the main agricultural sources producing greenhouse gas emissions.[86] To reduce greenhouse gas emissions from agricultural production takes also part of the "menu of solutions" from the abovementioned WWR report. According to WWR report,[86] agricultural production was responsible for 6.8 Gt of greenhouse gas emissions in 2010 and will reach 9 Gt in the 2050 baseline scenario, of which they estimate GHG emissions from managed manure at nearly 590 million tons (Mt) of CO_2 in 2010, projecting to rise 770 Mt CO_2 in 2050. Of this total, they estimate that roughly one-third is in the form of nitrous oxide and two-thirds is in the form of methane. They propose as one of the recommended strategies to address this issue, to adopt competitive programs to encourage new technology, which can build upon waste-treatment technologies already developed for industrial wastes and municipal sewage. In this line of waste treatment, plasma technology has already come a long way, having been implemented for several years in treatment facilities for different types of waste.[174]

C. Plasma Activated Water. A way to scale-up plasma treatments

As has been already described, the use of plasma technology in agriculture sometimes is required to be applied *in situ* in the growing area. This would be the case for example of soil or crop treatments when it comes to dealing with plant diseases generated by microorganisms or insects. The main difficulty of plasma application for these cases is its necessary scaling to treat large areas of cultivation. One of the ways to deal with escalation would be the use of plasma activated water (PAW), which can be considered as a simple means for transmitting the plasma action (or part of it). Indeed, more than a decade ago it was described that the interaction of a plasma with water triggers a series of chemical reactions and physical phenomena that result in the generation of certain species reactive species (including reactive oxygen species and reactive nitrogen species), and in the change of some water properties such as its pH and conductivity.[175] Kamgang-Youbi et al reported on the bactericidal action of such activated liquids,[176] governed by mechanisms similar to those inducing decontamination in CAP containing water, and coined the term "plasma-activated water". Since then the possible uses of this activated water (and other activated liquids) have been investigated, although disinfection has been mainly reported. Compared to plasma, the action of PAW would be milder, as it would avoid surface effects such as etching. Two different comprehensive reviews on its physicochemical properties, antimicrobial effectiveness, and microbial inactivation mechanisms, including applications in agro-food industry have been recently published,[177,178] thus here, we will just give some hints about its usefulness as a method to solve the plasma scaling problem in some of the applications in agriculture.

Park and coworkers reported for the first time in 2013 on the effect of plasma activated water treated with different plasma sources on plant development and showed PAW allows increasing the growth of plants, enhancing the action of fertilizers.[179] The capture of atmospheric nitrogen in water from plasma treatment turns water (to a greater or lesser extent, depending on the treatment performed) into fertilizer. The ability of PAW promoting germination and seedling growing was confirmed in subsequent studies published.[180-184] Therefore, the use of PAW in crops would be a good and sustainable strategy to promote yield while reducing the use of chemical fertilizers. Moreover, Zambon and colleagues have recently demonstrated PAW treatment enhances the plant defense responses, and proposed its use as immunity inducer under greenhouse conditions to increase the yields and crop quality reducing the water usage,[152] which would further reduce the consumption of fertilizers.

On the other hand, because the disinfectant action of plasma-activated water is obvious its usefulness for the control of pathogens and animal pests in crops. The main advantage of this practice in the control of disease dissemination is its sustainability as it would avoid the use of pesticides, helping to reduce the amount of agrochemical delivered to the environment and limit one of the biggest problems today, which is that of pathogen and animal resistance.

Finally, PAW can be also used as a washing disinfectant for food products, including fruits and vegetables, meat, and seafood. Freshly cut fruits (apple, kiwi, pears), grapes, Chinese blueberries, strawberries, baby spinach leaves, iceberg lettuce, romaine leaves ... are some of the fresh products for which plasma disinfection (bacteria, mold, yeasts, fungi) has proven its effectiveness. This type of treatment is very useful as it would lead to an extended shelf-life.

Although today the use of PAW is even commercialized,[185] certain aspects concerning its properties and effects of its use (especially when applied directly to food) still need to be clarified. In this way, a better knowledge of the different oxidizers formed in PAW and a better understating of the mechanisms leading to microbial inactivation process is still needed, which allow preparation of PAW with adequate properties to address determined plant diseases. Studies of chemical changes and toxicity on food components upon PAW interaction are still scarce and necessary since they will allow adjusting the doses and the way of application of these treatments.

V. CONTROLLING THE SPREAD OF INFECTIOUS DISEASES

A. Challenges in controlling the spread of infectious diseases

With the emergence of the COVID-19 outbreak, in recent months, we have witnessed an unprecedented event which has highlighted the need to develop robust protection and prevention strategies to improve disease control mechanisms. But, prior to the COVID-19 outbreak, there were also important pending tasks related to the spread of infectious diseases awaiting to be addressed, as it is the case of healthcare-associated infections (HAIs). HAIs are one of the most common adverse events affecting patient safety worldwide.[186] These infections, occurring in a patient while receiving care in a hospital or other healthcare facilities, are mainly caused by microorganisms resistant (and in many cases multi-resistant) to commonly used antimicrobials. Although most prevalent in low- and middle-income countries, all countries currently have a burden caused by HAIs.[187] Due to the difficulty in collecting reliable data, it is not possible to exactly know the global burden caused by this type of infections, but it is estimated that hundreds of millions of patients are affected by HAI each year (around 2 million patients in the US and 3 million patients in the EU)[188] which, in addition to putting into risks the health of professionals and patients, generates significant morbidity and mortality, as well as financial losses for patients and health systems. The identification and recognition of the severity of this problem have led in recent years to an intensification of research on the pathogenesis and management of this type of infection, and the development of new prevention approaches. Surveillance and prevention are key elements in solving this serious issue, as most HAIs are thought to be preventable.[188]

Between 80% to 87% of HCAIs are caused by 12 to 17 microorganisms including *S. aureus*, *Enterococcus spp.*, *E. coli*, *P. aeruginosa*, *Enterobacter spp.*, *Klebsiella spp.*, coagulase negative *staphylococci*, *Candida* species and NOS yeasts.

Among HAIs, Surgical Site Infections (SSI) have a relatively high prevalence (despite being one of the most preventable). According to the World Health Organization,[187] SSI is the most common type of HAI in low- and middle-income countries (affecting up to a third of patients who have undergone a surgical procedure), and remains the second most common HAI in Europe and the

United States of America. There is also a perception among the public that SSIs can reflect poor quality of health care. For these reasons, SSI prevention has received considerable and increasing attention in recent years.

B. The approach

According to recent evidence, a contaminated healthcare environment plays an outstanding role in the transmission of microorganisms and, in this sense, the document "Global guidelines for the prevention of surgical site infections" published by the WHO points out the importance of a clean environment in operating rooms, as well as of the decontamination of medical devices and surgical instruments to prevent SSI.[187] Adequate cleaning and disinfection in the operating room of hand-touch surfaces and surfaces contaminated with blood and body fluids, and correct sterilization of surgical instrumentation is essential to avoid the spread of infections.

On the other hand, the appropriate and rational use of air purification methods improves the quality of the air in the operating rooms and contributes to reducing the risk of SSI.[189] This is particularly critical when microbial transmission via aerosols is implied. In China, the number of hospitals using air cleaning systems in operating rooms has markedly increased in recent years, as this cleaning strategy is considered relevant in the control of SSI. Thus, the so-called Air Cleaning Technology (ACT) and other air purification methods including ultraviolet, light disinfection, circulating wind UV sterilizer, and electrostatic adsorption air sterilizer, have been widely introduced in hospitals and hospital operating rooms in China. However, the right selection and proper use of the different air purification methods still need to be optimized.

The abovementioned prevention strategies acquire a special relevance in the case of Intensive Care Units, whose patients are especially susceptible to HAIs because they are often in an immuno-compromised status. It is estimated that HAIs can affect 9% to 37% of patients in intensive care units.[186]

Adequate management of waste derived from health care is also crucial for the control of disease transmission, as waste often acts as reservoirs for pathogenic microorganisms. In general, not only does contagious waste management matter, but also coping with pathological, sharp, chemical, medicinal, cytotoxic, radioactive, and other non-hazardous wastes. In recent years, the generation of medical waste has been growing at rates higher than the infrastructures can manage.[190] According to WHO data, prior to the outbreak of the COVID-19 pandemic, high-income countries generated on average up to 0.5 kg of hazardous waste per hospital bed per day, while low-income countries generated an average of 0.2 kg.[191] These numbers should have notably risen in the last year and a half, a period in which the COVID-19 pandemic has caused an unforeseen collapse of waste management chains. You et al have reported a massive increase of medical waste in Wuhan (China) from between 40 and 50 tons/day before the outbreak to about 247 tons on 1 March 2020.[192]Although in recent years, the disposal of hazardous medical waste has been regulated with increasingly strict regulations in many countries, this pandemic has further alleviated the need for safe and rapid methods of managing this type of waste.

Inadequate treatment and disposal of healthcare waste can indirectly pose health risks through the release of pathogens and toxic pollutants into the environment.[191] The disposal of untreated sanitary waste in landfills can lead to contamination of drinking and ground waters. The treatment of sanitary waste with chemical disinfectants can cause an eventual release of environmentally polluting substances if there is not environmentally sound handling and disposal. Waste incineration is still widely practiced, but improper incineration can result in the release of dangerous pollutants into the air and the generation of ash residues. Thus, the incineration of materials with a high metal content (in particular lead, mercury and cadmium) can cause the spread of toxic metals in the environment. Also, the incineration of materials containing or treated with chlorine can generate dioxins and furans, secondary pollutants that are carcinogenic and associated with other serious health issues. The search for alternatives to incineration is therefore necessary. WHO states that healthcare waste management requires increased care and diligence to avoid adverse health outcomes associated with poor practices.

C. Opportunities of non-thermal plasmas in controlling the spread of diseases

In the context of prevention for disease control, the use of plasma technology can be a powerful and effective strategy. In fact, NTAPs have proven to be a very efficient tool to disinfect surfaces and sterilize medical instruments, in the decontamination of air currents, as well as in the management of medical waste. On the other hand, from the point of view of clinical practice, due to its antiseptic action, recent studies have shown the efficacy of plasmas in wound healing, allowing to reduce the use of antibiotics and thus helping to limit the development of resistance of microorganisms.

In 1968, Menashi reported for the first time the efficacy of plasma as a sterilizing agent.[193] Sterilization/decontamination was the first application of plasmas in the medical field and is today the best known. The biocidal activity of plasma is mainly based on the oxidative action of some of its species (including O_3, hydrogen peroxide, NO, NO_2, photons) and its bio-decontaminating/sterilizing efficacy has been repeatedly demonstrated.[194-197] Since then, research in this field has been extensive until today, using both atmospheric pressure and reduced pressure plasma reactors.[198-246] Plasmas have been shown to be effective in the decontamination/sterilization of bacteria, biofilms, spores, fungi, and viruses. Oxidative damage to microorganism cell membranes, proteins, enzymes, DNA damage, mitochondria dysfunction, among others, have been reported as leading to cell death. However, despite the advanced research in this field, there is still no commercialized sterilizing device based on the use of plasma technology. Beckeschus and colleagues have attributed that to the strong requirements imposed by the pharmacopeia and other regulations to medical sterilization procedures.[196]

Nevertheless, the COVID-19 outbreak has created an unprecedented situation that has highlighted the urgent need to develop new methods to interrupt the chain of viral infection and mitigate the pathogenesis associated with SARS-CoV-2 (and, so, potential new future viruses). In this sense, it is worth highlighting the intensification in research on plasma-assisted sterilization during the last year, having even already analyzed the efficacy of cold plasmas against this new coronavirus and virus in general.[239,242] We believe that this new stage provides an opportunity for plasma technology to establish itself as a complementary sterilization tool in the field of medicine. Once this technology is implemented, it will also be very useful in the elimination of microorganisms from surfaces and medical instruments (some already resistant to conventional sterilization methods), helping to cope with the important challenge of HAIs and SSIs. *Escherichia coli*, *Klebsiella pneumoniae*, *Enterobacter aerogenes*, *Enterobacter cloacae*, *Enterobacter faecalis Staphilococus aureus*, *Staphilococus epidermidis*, *Bacillus subtilis*, *Streptococcus salivarious*, *Streptococcus pneumoniae*, *Streptococcus pyogenes*, *Candida albicans*, hepatitis A virus, respiratory syncytial virus, herpes simplex virus… are among microorganisms against which plasmas have been shown to be effective. Let us remark that this technology is already developed, the only thing left is to use it.

Plasmas are also efficient in air bio-decontamination, thus helping to create safer environments. One of the first devices allowing air bio-remediation was the so-called Pathogen Detection and Remediation Facility designed and built at the Drexel Institute in Philadelphia by Vaze and colleagues using a dielectric barrier grating discharge.[195,247] However, this type of application had also not been used much in practice and, until the COVID-19 outbreak. Prior to pandemics only a few air purification systems based on plasma technology had been commercialized.[154,248] Once aerosols were identified as the main route of transmission of SARS-CoV-2 the last year, the presence of plasma technology in the market of air purifiers has remarkably grown, and thus we can found some new systems such as *Covidinator*,[249] *Plaze Safe Air Disinfector*,[250] *Plasma Ozone Air Disinfection*,[251] *Aire eco3 Hispacold*,[252] to name just a few. Most of them, are based on the so-called Needlepoint Bipolar Ionization (NPBI™) technology, developed by GPS company, relying on the use of a plasma to generate high concentration of negative and positive ions, as well as some RONS.[253] Plasmacluster® air purification technology developed by Sharp Corporation in 2000,[254] is another plasma approach based on generation of oxidizing species. Sharp claims that, recently, researchers at Nagasaki University and Shimane University in Japan have proven that this device reduces the concentration of aerosolized SARS-CoV-2 passing through a test chamber by 91% after 30 s of exposure.[255] Currently, the lack of more rigorous peer-reviewed studies on this emerging technology in the air conditioning and cleaning sector is responsible for the fact that environmental protection agencies do not yet propose it as efficient and safe.[256] And it would be worth it to encourage multidisciplinary research on this topic, proving its high efficiency in terms of microbial deactivation, as it is a sustainable technology not using toxic chemicals.

Plasmas can also offer a valuable and safe solution to the treatment of medical waste. For this purpose, thermal plasmas are needed. Although they fall outside the purpose of the article, we will give a brief overview of their usefulness in this matter. Thermal plasma technology for the treatment of solid waste is already commercialized being used today for the elimination or transformation of different types of waste (urban solid waste, electronic waste, steelmaking waste, electroplating waste, asbestos, plastic waste, biomass, radioactive waste…).[257-261] Thermal plasma reactors can be used to melt and vitrify waste into a stable, non-leachable glass slag, which can be harmlessly disposed of in an inert landfil and even sometimes advantageously reutilized. Also, by gasification or pyrolysis, plasmas can transform the organic fraction of the waste into a hydrogen-rich synthesis gas.

The use of thermal plasma reactors has, in general, unique advantages in the destruction of hazardous waste, which include (i) their ability to treat a wide range of waste (solid, liquid and gaseous); (ii) they have a high energy density and temperatures that lead to fast reaction times and high throughput; (iii) they do not require the addition of oxidizing agents, (iv) they allow metal recovery, (v) or the possibility of being integrated into the process that generates the waste. And all this with none or minimal production of dangerous secondary substances such as furans and dioxins.[191,255] In addition to this, thermal plasmas can fully destroy any microorganism given their extremely high temperature, which could be of great importance in the management of medical waste.

Interestingly, the existing commercial facilities for the treatment of waste based on the use of plasmas offer the possibility of managing all types of waste, except medical waste. This could be due in part to the difficulties associated with handling this type of waste during transport to, as well as within, the treatment plant. On the other hand, although treatment with thermal plasmas undoubtedly eliminates any microorganism, given the obviousness, there are also only a few scientific studies that demonstrate the effectiveness of these specific reactors for biodegradation,[262,263] which makes it difficult to sell this technology for that end.

But, the ability to perform on-site waste treatments in hospitals, offering a proximal solution for better control of biologically contaminated waste, is one of the great attractions of plasma technology, which has yet to be exploited. Indeed, although research on medical waste treatment has been extensive through the design of pilot plasma reactors and the use of surrogates to simulate human tissues and organs, this technology has not been implemented so far in any hospital or medical facility. This fact may also be a consequence of the structural failures that, according to the WHO, medical waste management in general still has today, including the absence of waste management and disposal systems, insufficient financial resources and humans and the low priority given to this issue.

Concluding remarks

As can be seen from the anecdotes presented here, while not exhaustive, plasmas offer potential solutions to a wide range of environmental problems. Indeed, this perspectives piece did not cover plasma catalysis directly though this field also offers a host of solutions to environmental problems as well such as carbon dioxide decomposition, fuel reforming, and nitrogen fixation—all key engineering challenges.[264] Atmospheric pressure plasmas offer unprecedented, nonequilibrium control of gas phase processes that drive reactivity all at atmospheric pressure—thus simplifying implementation. In many cases discussed, the status of these approaches are at the early-stage—essentially bench scale demonstrations. Perhaps one of the biggest challenges to realizing these solutions is demonstration of scalability within the bounds of economics constraints when compared to conventional technologies used for the same purpose. Continued research is certainly warranted as the potential benefits if realized can literally be transformative.

We have analyzed the way that plasmas can contribute to the challenge of clean water for all. Addressing water scarcity derived from climate change will be a major challenge in the coming decades. The occurrence of emerging pathogens in water, the threat posed by both inadequate removal of industrial toxins in drinking water (leading to long term health problems, including cancer) and pharmaceuticals often dispensed in waste streams (contributing to aggravating the important issue of antibiotic resistances), among others, highlight the need for advanced treatment methods and water management policies. Plasma-based water purification offers a compelling solution to a host of water quality problems. The non-selective and indiscriminate character of the physical mechanisms of decomposition in plasmas make them very adequate to address a range of contaminants including those accidentally introduced or those that are currently emerging. Nevertheless, from a chemical perspective, there is still much to be learned regarding the decomposition pathways driven by plasmas, being particularly critical to understand the degree of mineralization and the level of toxicity of intermediates formed during processing.

Plastic waste is another looming environmental issue facing society today. To cease the production of new plastics and recycle the waste plastic by depolymerization and generate new products through the upscaling of the resulting monomers is the ultimate solution to this problem. Early experiments suggest that plasmas may indeed be able to play a role in such recycling. The potential promise of plasma-based processing of plastics relies on the fact that plasma driven process is completely non-thermal, residing the bulk of the reactants and solvent solution at room temperature, so being the energy loss is minimal. Additional investigation in the area of non-thermal plasmas in terms of selectivity and control allowing for optimization of scission leading to high yield depolymerization is still needed.

In the area of agriculture and food, the applications of non-thermal plasma processes are in the early stages of their investigation. This is a highly interdisciplinary topic that still has large margins of improvements and a long way to go in understanding and optimizing the fundamental processes and effects, before leaving the laboratory. Questions like scalability, efficiency, and consumer safety remain to be addressed. Comparison of non-thermal plasma technology with alternative approaches (in terms of performance and energy consumption) in some of these applications (e.g. plant growth) is often complicated due to the complexity to fix environmental conditions.

Finally, in the context of preventing and controlling the spread of disease, the use of plasma technology can be a powerful and effective strategy due to its efficiency in inactivating multi-drug-resistant pathogens. The implementation of this technique, however, has to overcome the challenge of achieving the accreditation of plasma-based processes and procedures. Due to these regulatory reasons, in the near term, plasma processes will most likely not replace classical sterilization processes, however, they will have a real chance of being used for specific decontamination when classical procedures are not effective. For this to happen, it has to be shown that the application of this technique does not lead to the development of resistance.

All in all, plasmas offer great potential for addressing a range of environmental and health challenges of direct benefit for society as a whole. Successful implementation however requires a good understanding of the problem, the shortcomings of conventional methods, and an appreciation of the challenge of scaleup both from an architecture of implementation standpoint as well as a point of view that includes costs to operate. Scaleup remains s key challenge and thus represents undiscovered country with the potential for great societal benefit. Key to realizing scale up is an understanding of the plasma physics of implementing discharges at scale and the associated control of the plasma-induced gas phase and surface chemistry that ultimately translates the science into a technology.

ACKNOWLEDGMENTS

Portions of this work was supported by the U.S. Department of Energy, Office of Fusion Energy Sciences (Grant No. DE-SC0018058 and DE-SC0022189), the National Science Foundation (Award Nos. 1700848 and 1747739), the RACER Trust (Grant No. N029593), and the Anthropocene Institute. M.C. García thanks the FEDER program and the Spanish Ministry of Science and Innovation (PID2020-112620GB-I00 and PID2020-114270RA-I00 Grants).

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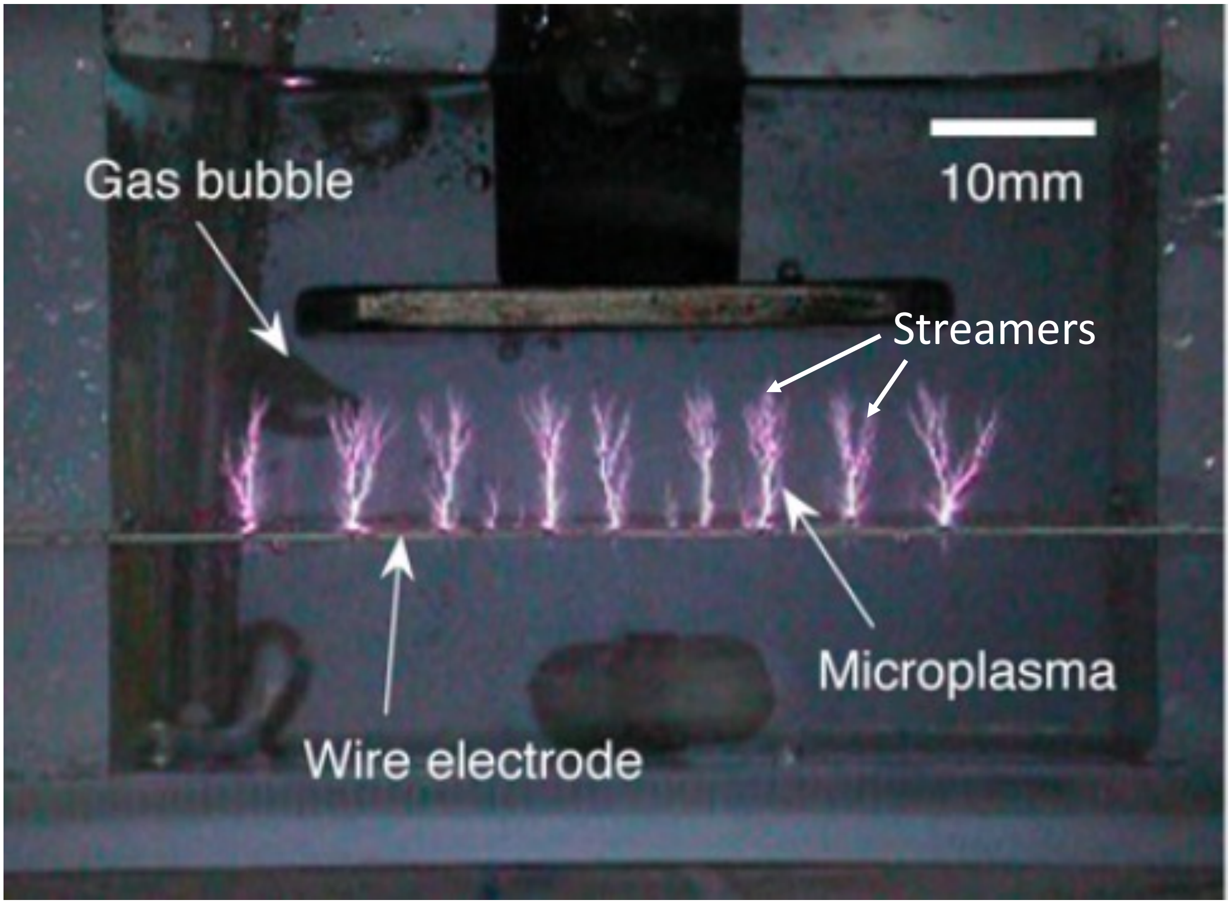
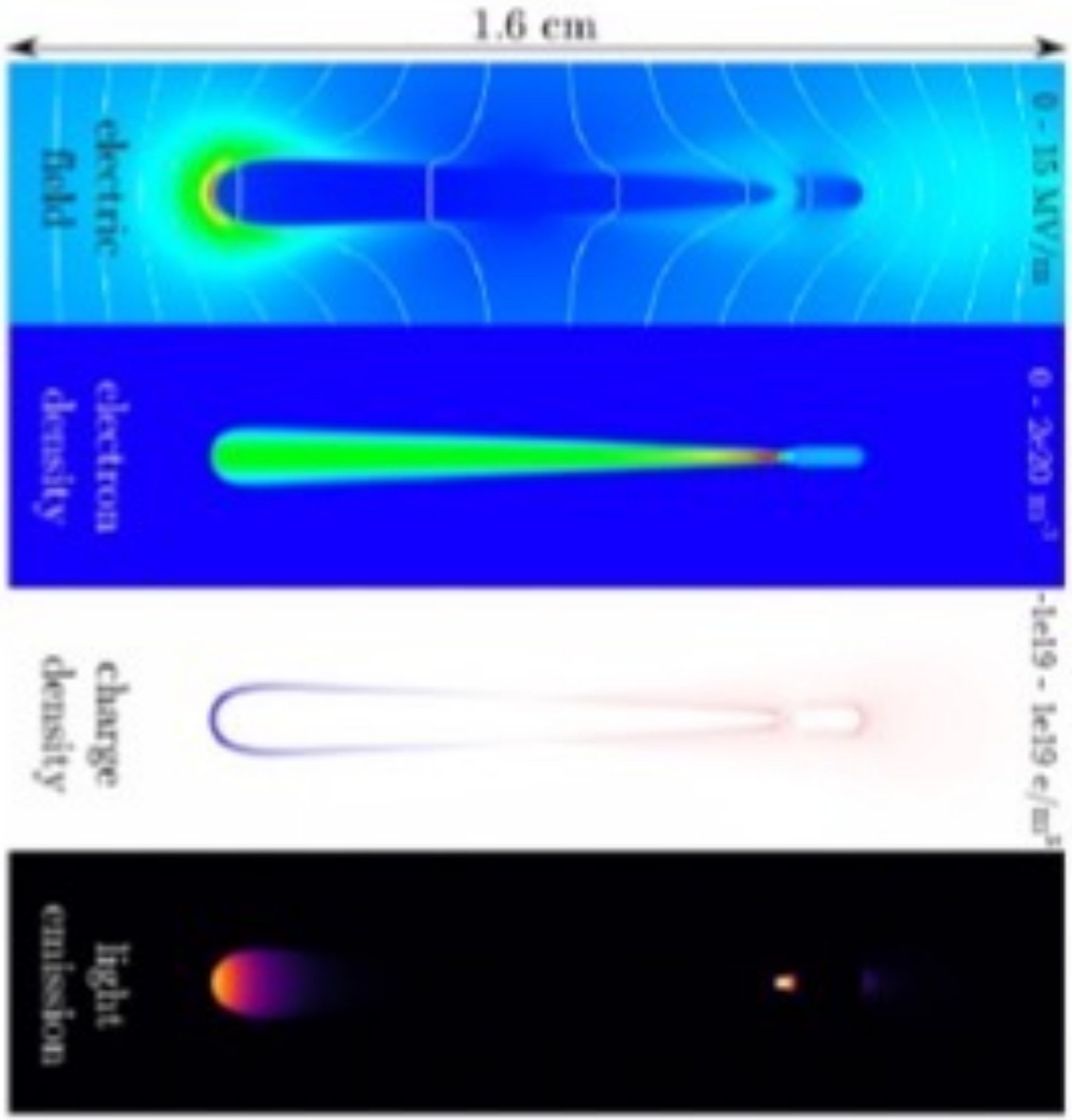
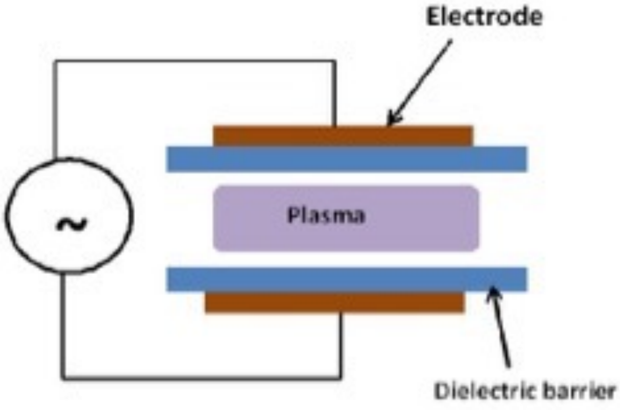
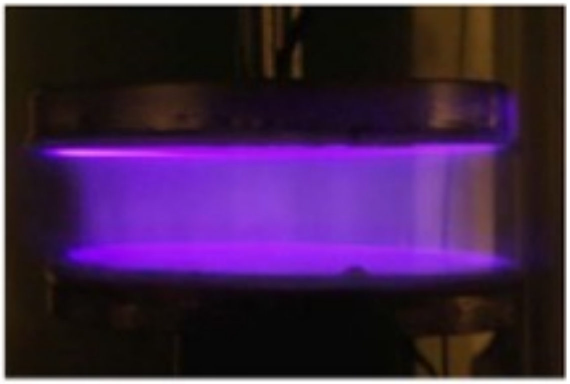

Gas bubble
10mm
Streamers
Microplasma
Wire electrode

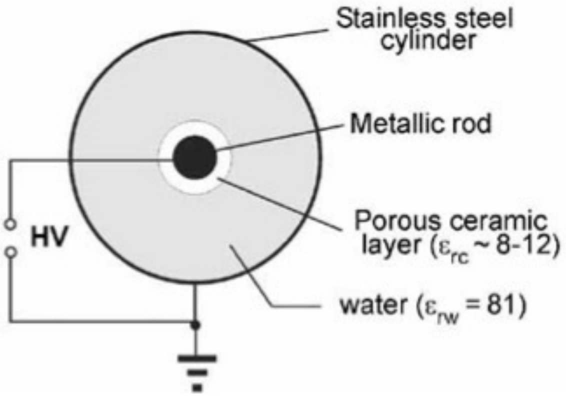

1.6 cm
0 - 15 MV/m
electric field
0 - 2e20 m⁻³
electron density
-1e19 - 1e19 e/m³
charge density
light emission

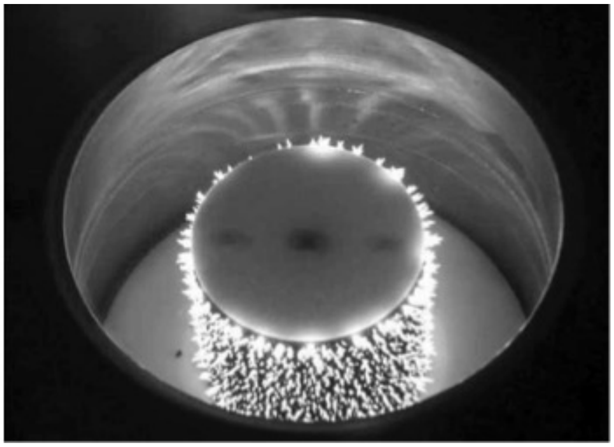


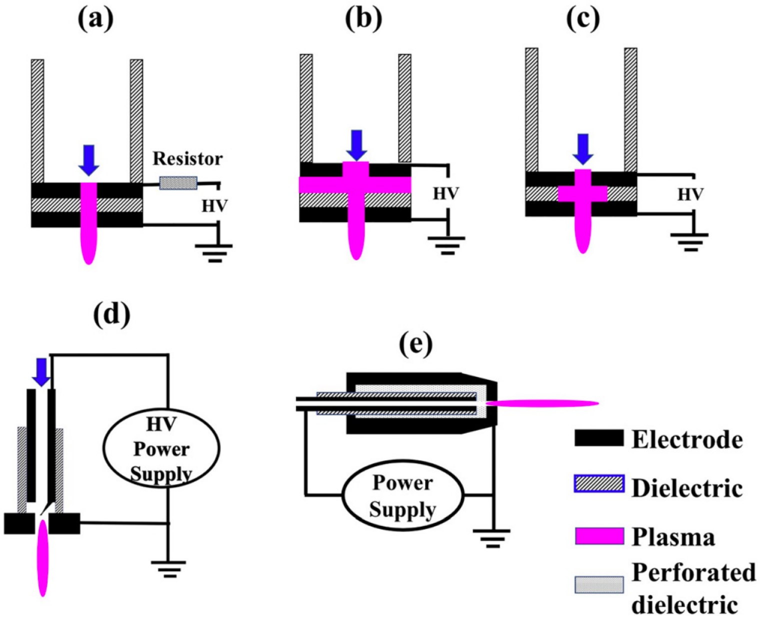
(a)

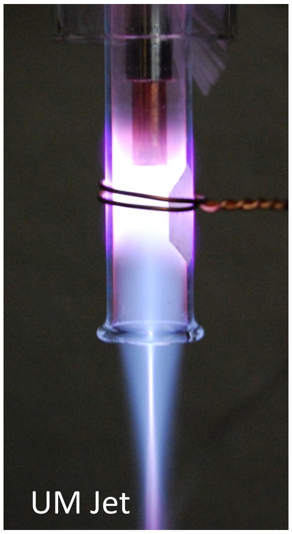


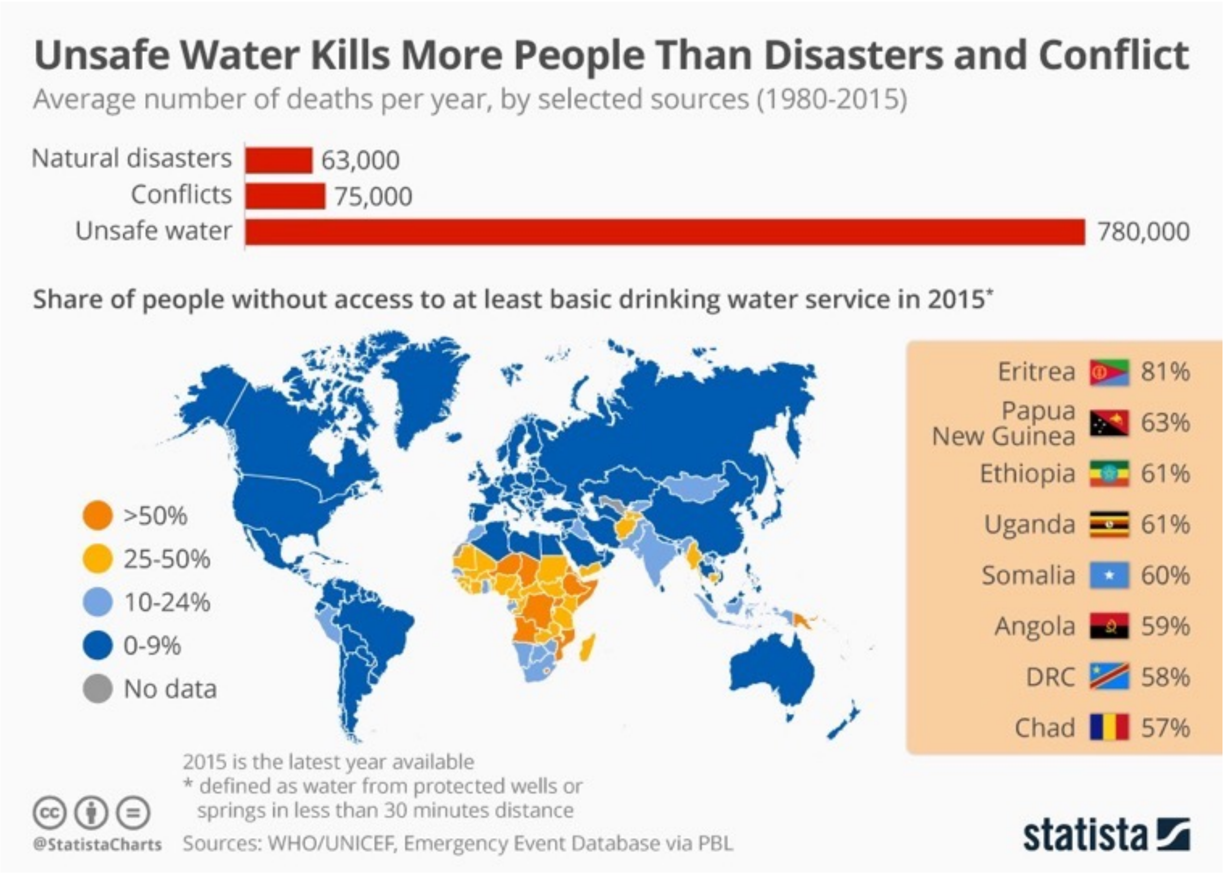
(b)


Stainless steel cylinder
Metallic rod
Porous ceramic layer (ε_{rc} ~ 8-12)
water (ε_{rw} = 81)
HV




(a)
Resistor
HV
(b)
HV
(c)
HV
(d)
HV Power Supply
(e)
Power Supply
Electrode
Dielectric
Plasma
Perforated dielectric


UM Jet


Unsafe Water Kills More People Than Disasters and Conflict
Average number of deaths per year, by selected sources (1980-2015)
Natural disasters
63,000
Conflicts
75,000
Unsafe water
780,000
Share of people without access to at least basic drinking water service in 2015*
>50%
25-50%
10-24%
0-9%
No data
Eritrea 81%
Papua New Guinea 63%
Ethiopia 61%
Uganda 61%
Somalia 60%
Angola 59%
DRC 58%
Chad 57%
2015 is the latest year available
* defined as water from protected wells or springs in less than 30 minutes distance
@StatistaCharts
Sources: WHO/UNICEF, Emergency Event Database via PBL
statista

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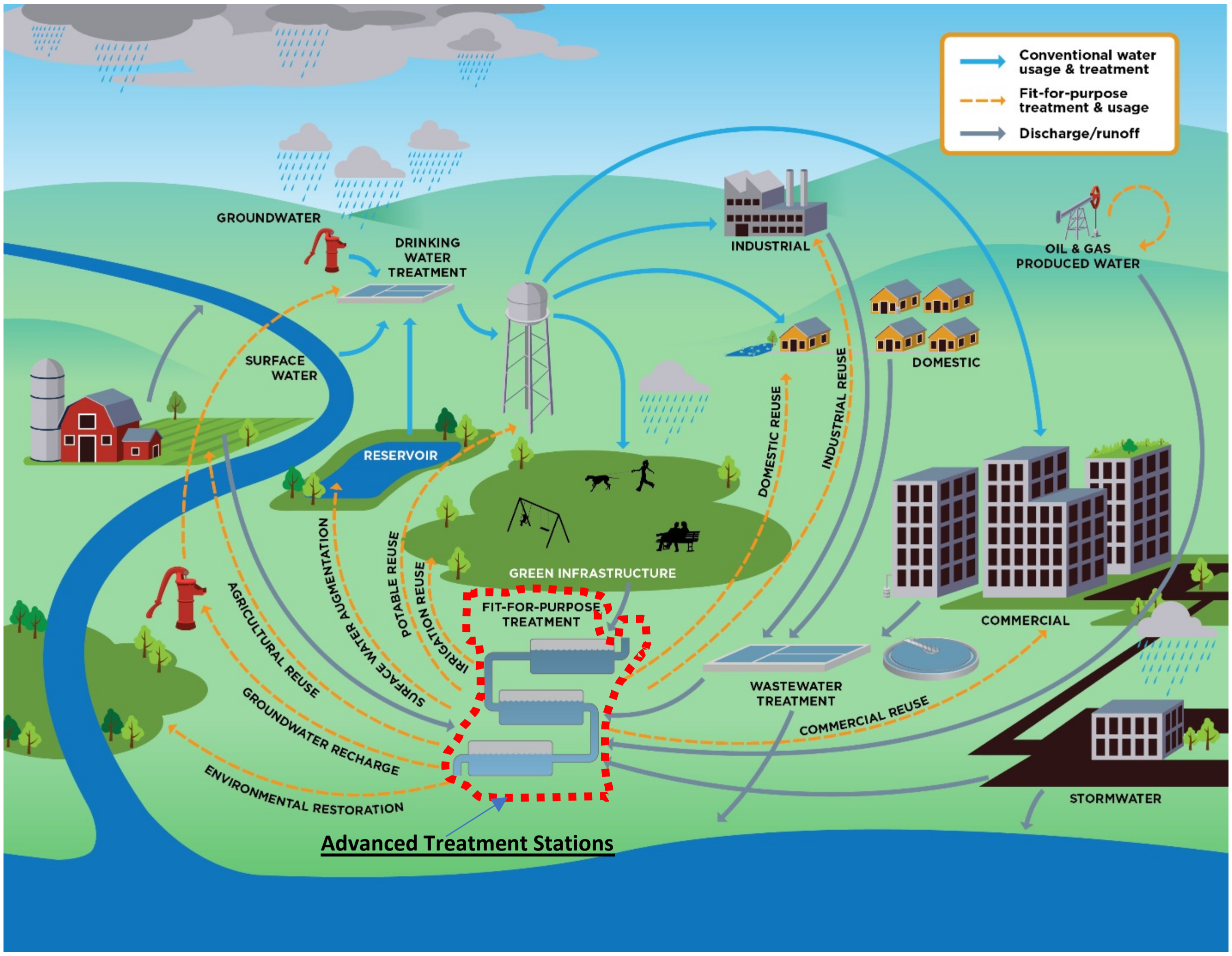
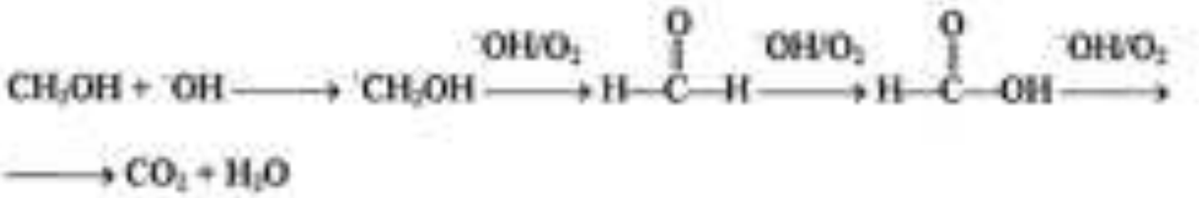
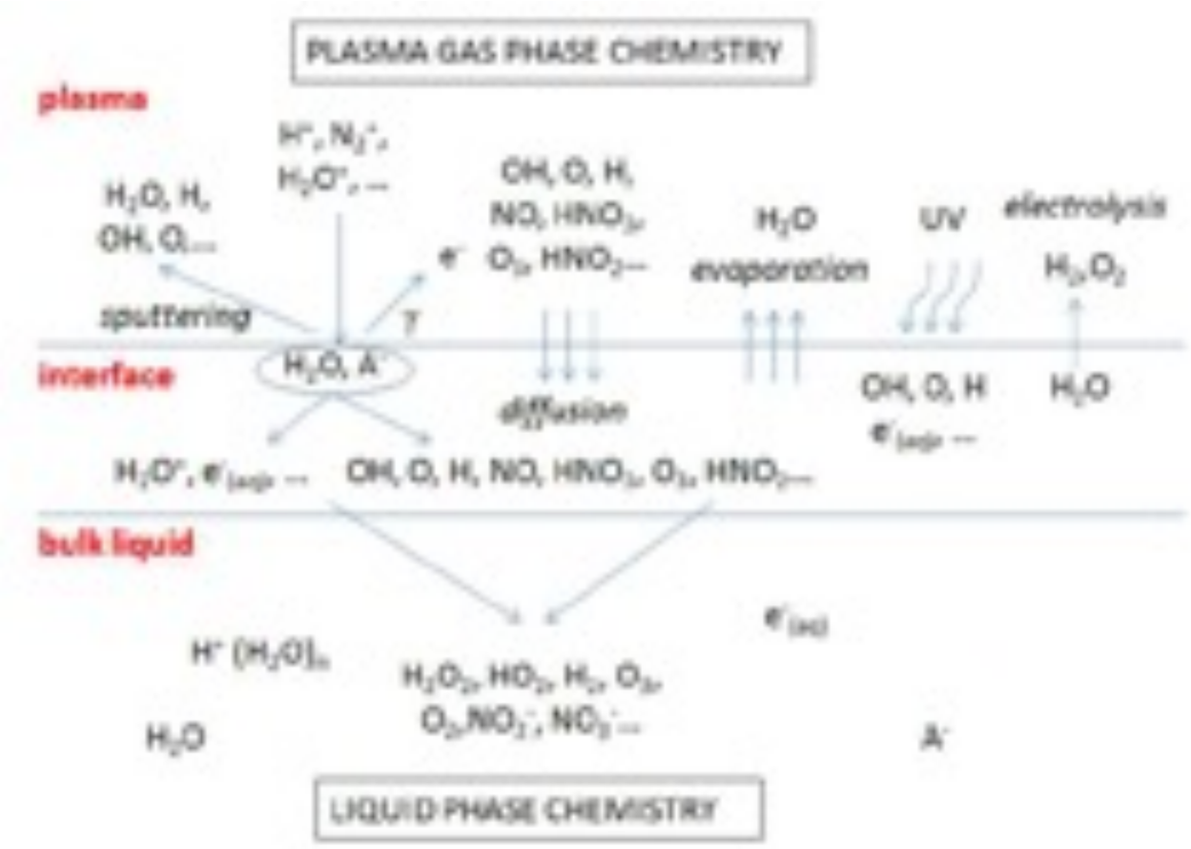
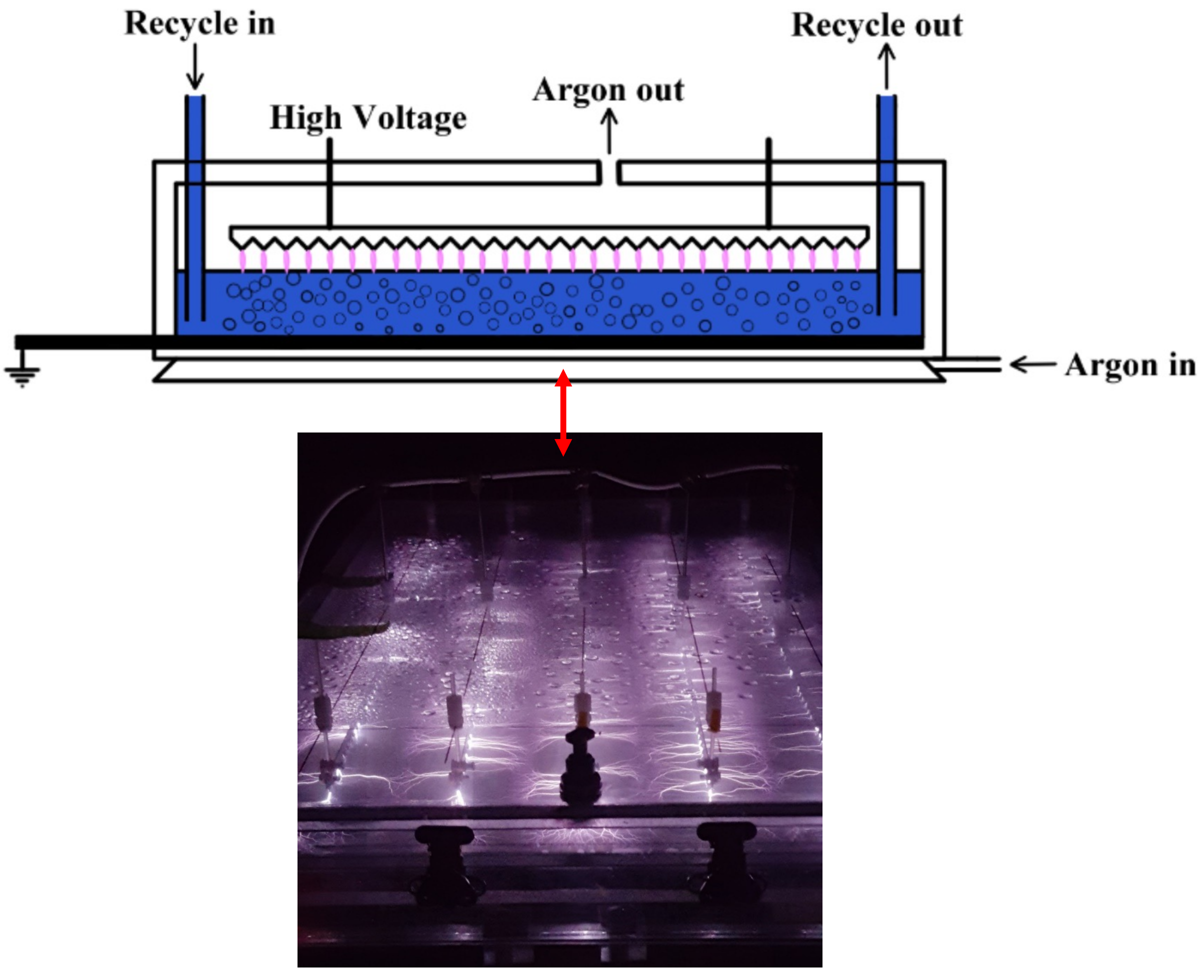
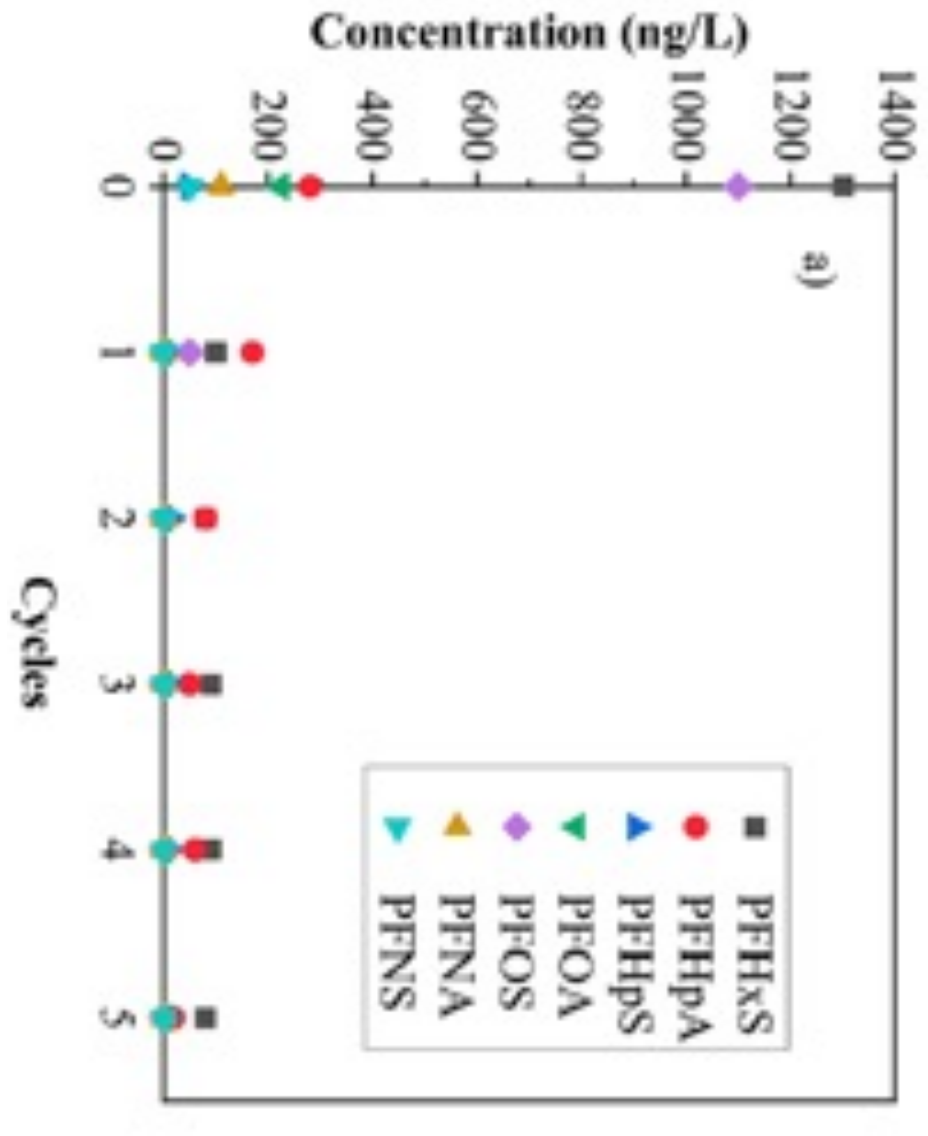

Conventional water usage & treatment
Fit-for-purpose treatment & usage
Discharge/runoff
GROUNDWATER
DRINKING WATER TREATMENT
INDUSTRIAL
OIL & GAS PRODUCED WATER
DOMESTIC
SURFACE WATER
RESERVOIR
DOMESTIC REUSE
INDUSTRIAL REUSE
GREEN INFRASTRUCTURE
COMMERCIAL
FIT-FOR-PURPOSE TREATMENT
AGRICULTURAL REUSE
SURFACE WATER AUGMENTATION
POTABLE REUSE
IRRIGATION REUSE
WASTEWATER TREATMENT
COMMERCIAL REUSE
GROUNDWATER RECHARGE
ENVIRONMENTAL RESTORATION
STORMWATER
Advanced Treatment Stations

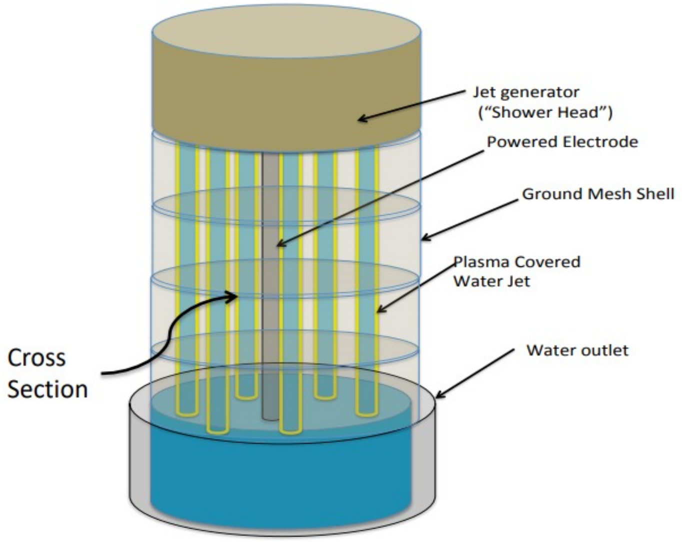


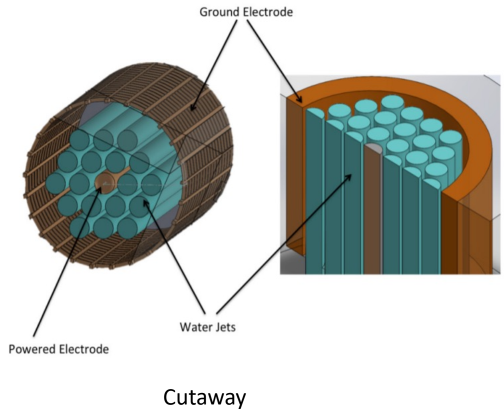
$$CH_3OH + {}^{\cdot}OH \longrightarrow {}^{\cdot}CH_2OH \xrightarrow{{}^{\cdot}OH/O_2} H-\overset{O}{\overset{\|}{C}}-H \xrightarrow{{}^{\cdot}OH/O_2} H-\overset{O}{\overset{\|}{C}}-OH \xrightarrow{{}^{\cdot}OH/O_2} \longrightarrow CO_2 + H_2O$$

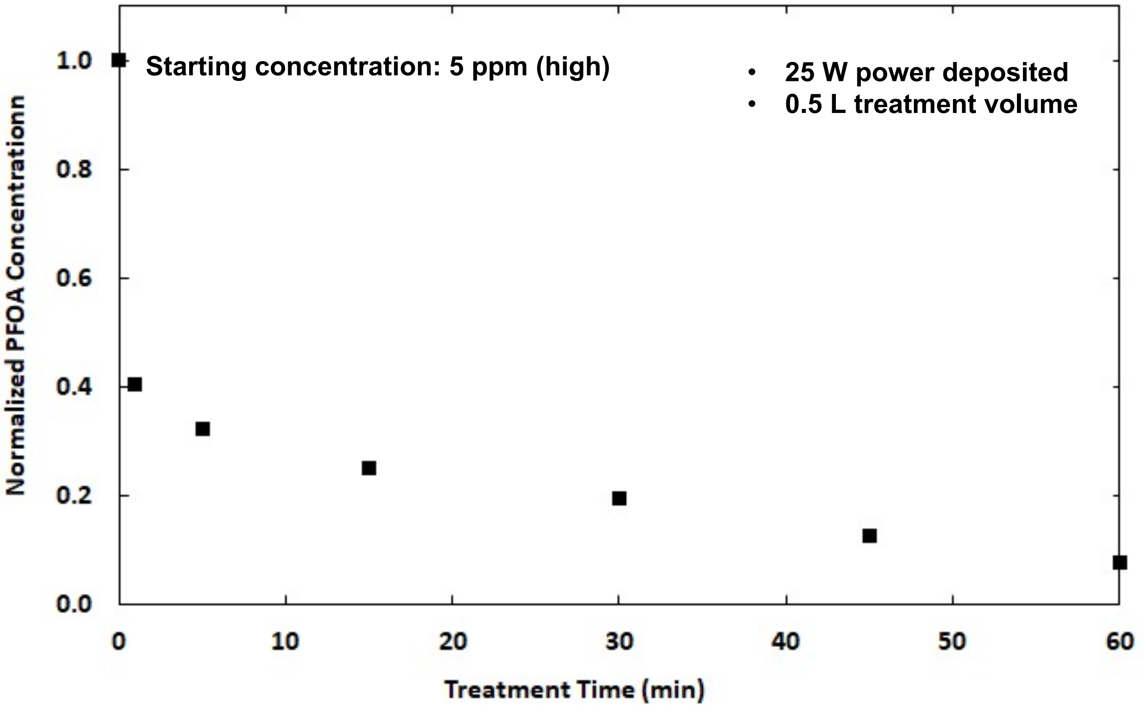

PLASMA GAS PHASE CHEMISTRY
plasma
H^+, N_2^+, H_2O^+, ...
H_2O, H, OH, O...
OH, O, H, NO, HNO_3, O_3, HNO_2...
e^-
H_2O evaporation
UV
electrolysis
H_2, O_2
sputtering
γ
interface
H_2O, A^-
diffusion
OH, O, H
$e^-_{(aq)}$ ...
H_2O
H_3O^+, $e^-_{(aq)}$ ...
OH, O, H, NO, HNO_3, O_3, HNO_2...
bulk liquid
$e^-_{(aq)}$
$H^+(H_2O)_n$
H_2O_2, HO_2, H_2, O_3, O_2, NO_2^-, NO_3^-...
H_2O
A^-
LIQUID PHASE CHEMISTRY


Recycle in
Recycle out
Argon out
High Voltage
Argon in


a)
Concentration (ng/L)
0
200
400
600
800
1000
1200
1400
Cycles
0
1
2
3
4
5
■ PFHxS
● PFHpA
▲ PFHpS
▼ PFOA
◆ PFOS
◀ PFNA
▶ PFNS


Jet generator
("Shower Head")
Powered Electrode
Ground Mesh Shell
Plasma Covered
Water Jet
Cross
Section
Water outlet


Ground Electrode
Powered Electrode
Water Jets
Cutaway


Starting concentration: 5 ppm (high)
• 25 W power deposited
• 0.5 L treatment volume
Normalized PFOA Concentrationn
1.0
0.8
0.6
0.4
0.2
0.0
0
10
20
30
40
50
60
Treatment Time (min)

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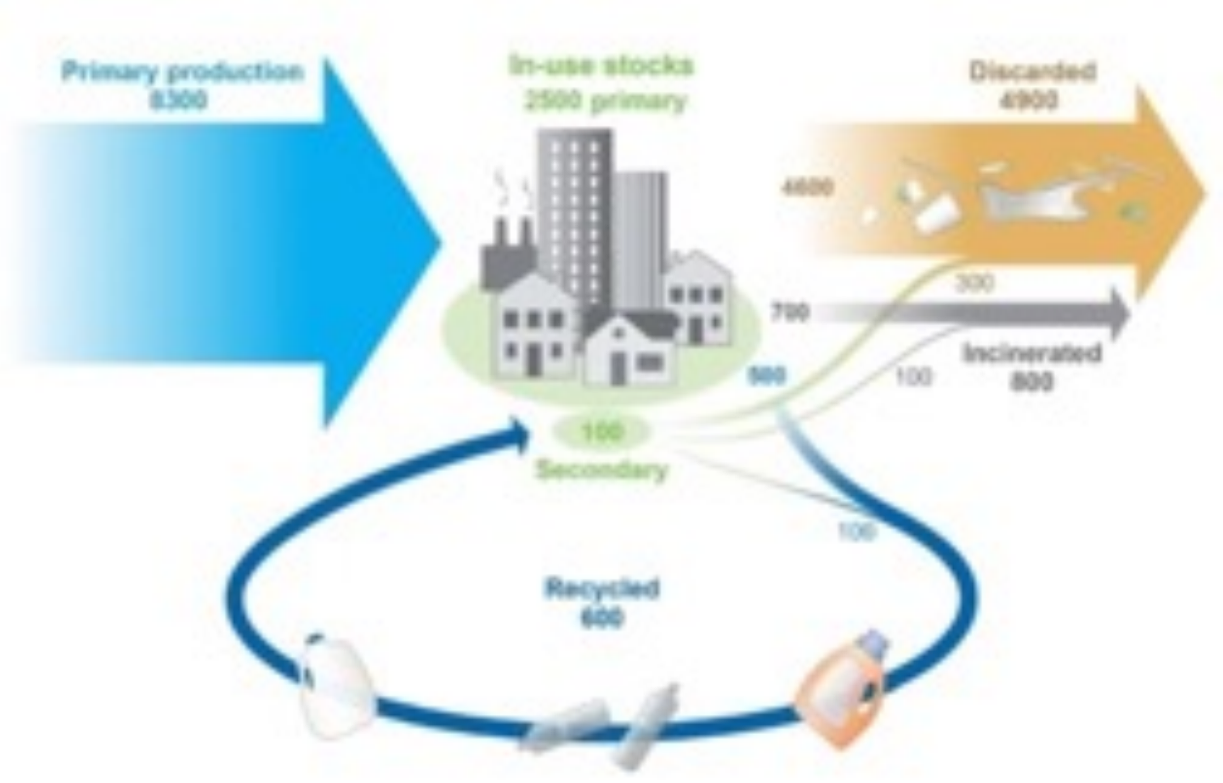
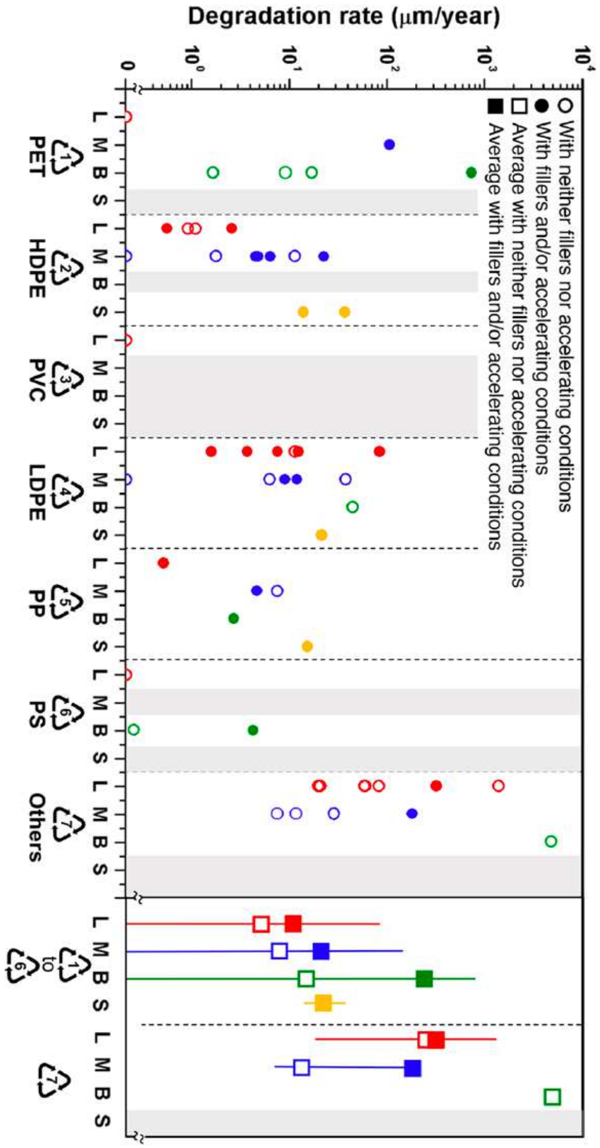
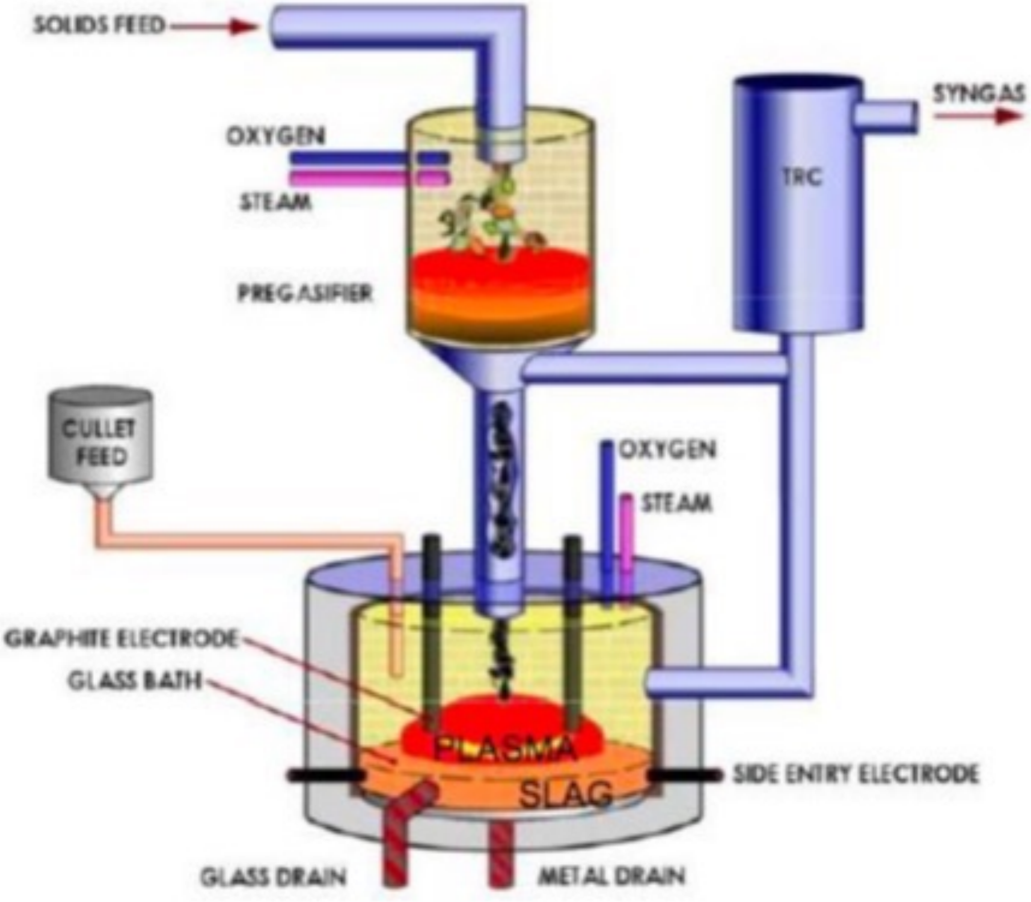



○ With neither fillers nor accelerating conditions
● With fillers and/or accelerating conditions
□ Average with neither fillers nor accelerating conditions
■ Average with fillers and/or accelerating conditions
Degradation rate (μm/year)
10^4
10^3
10^2
10^1
10^0
0
L M B S
1 PET
2 HDPE
3 PVC
4 LDPE
5 PP
6 PS
7 Others
1 to 6
7


SOLIDS FEED
OXYGEN
STEAM
PREGASIFIER
TRC
SYNGAS
CULLET FEED
OXYGEN
STEAM
GRAPHITE ELECTRODE
GLASS BATH
PLASMA
SLAG
SIDE ENTRY ELECTRODE
GLASS DRAIN
METAL DRAIN

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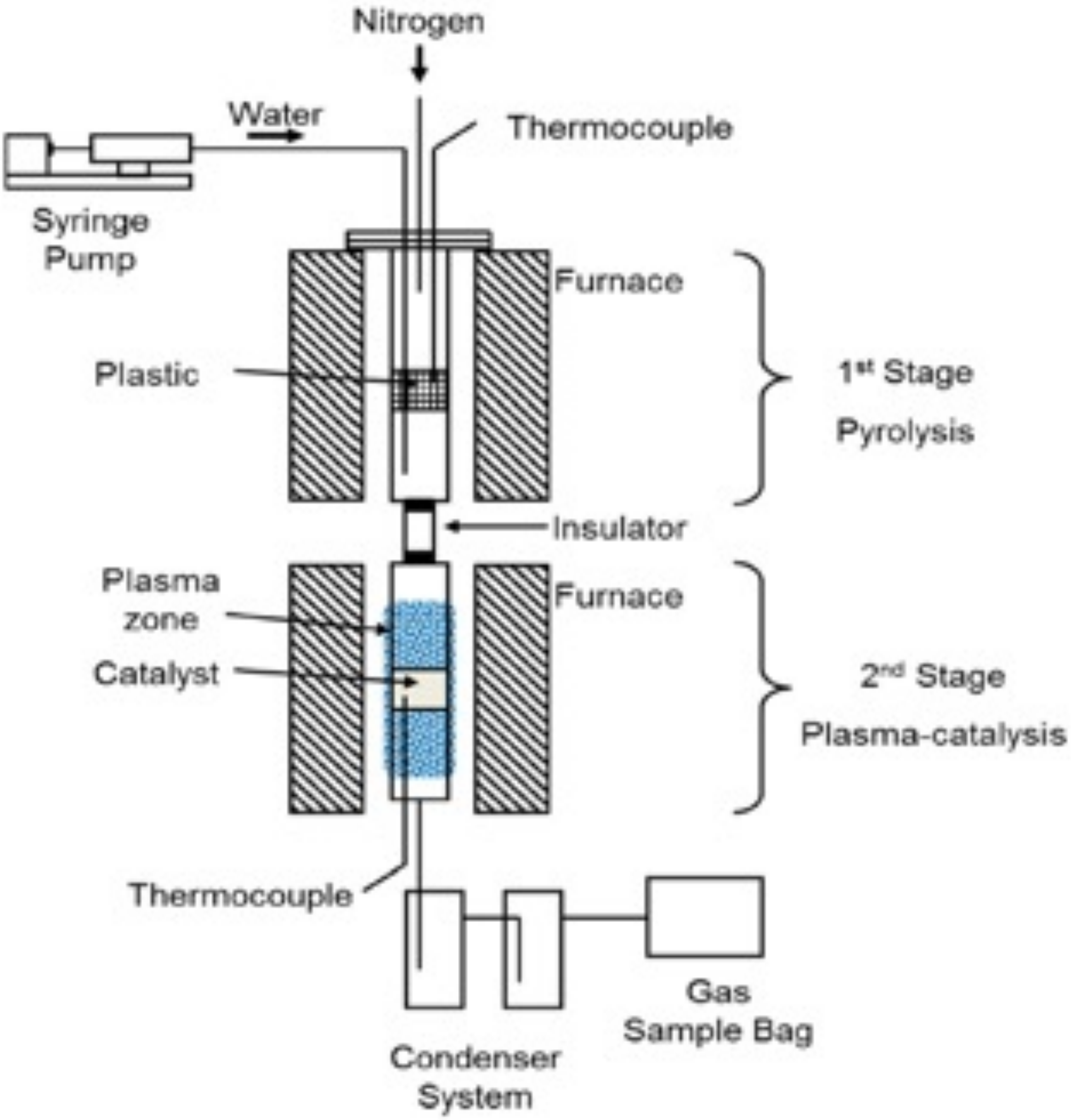

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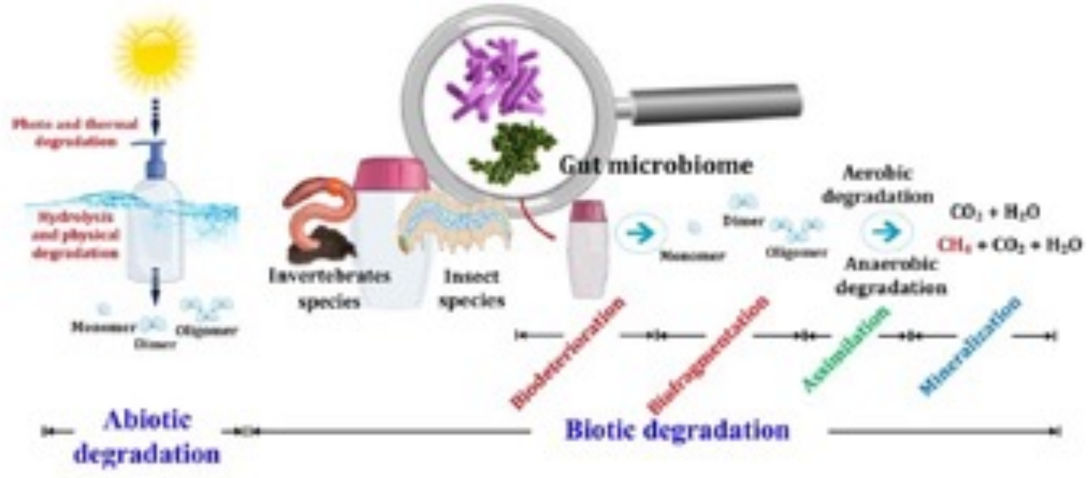

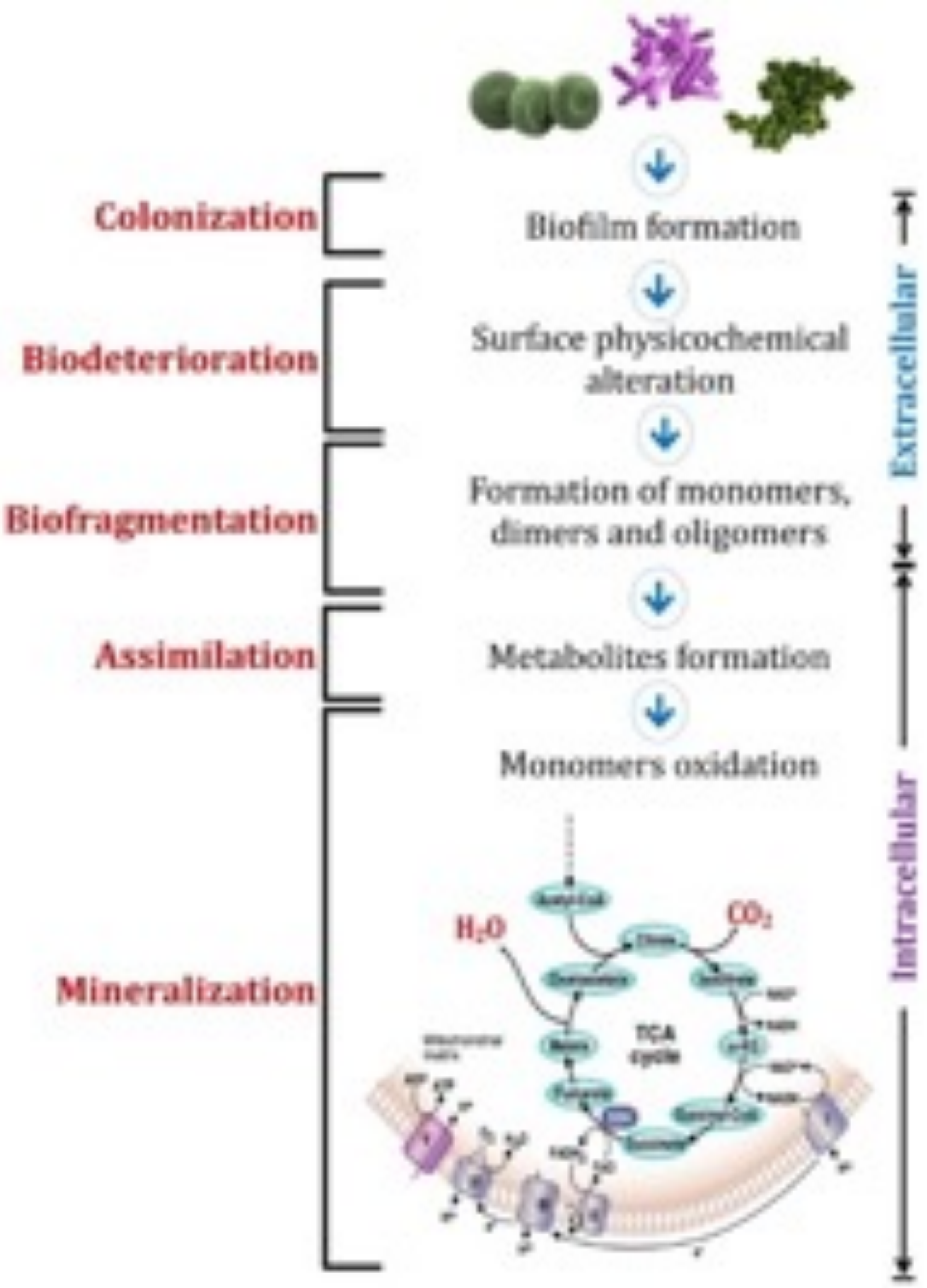
Colonization
Biofilm formation
Biodeterioration
Surface physicochemical alteration
Biofragmentation
Formation of monomers, dimers and oligomers
Extracellular
Assimilation
Metabolites formation
Monomers oxidation
Intracellular
Mineralization
H_2O
CO_2
TCA cycle

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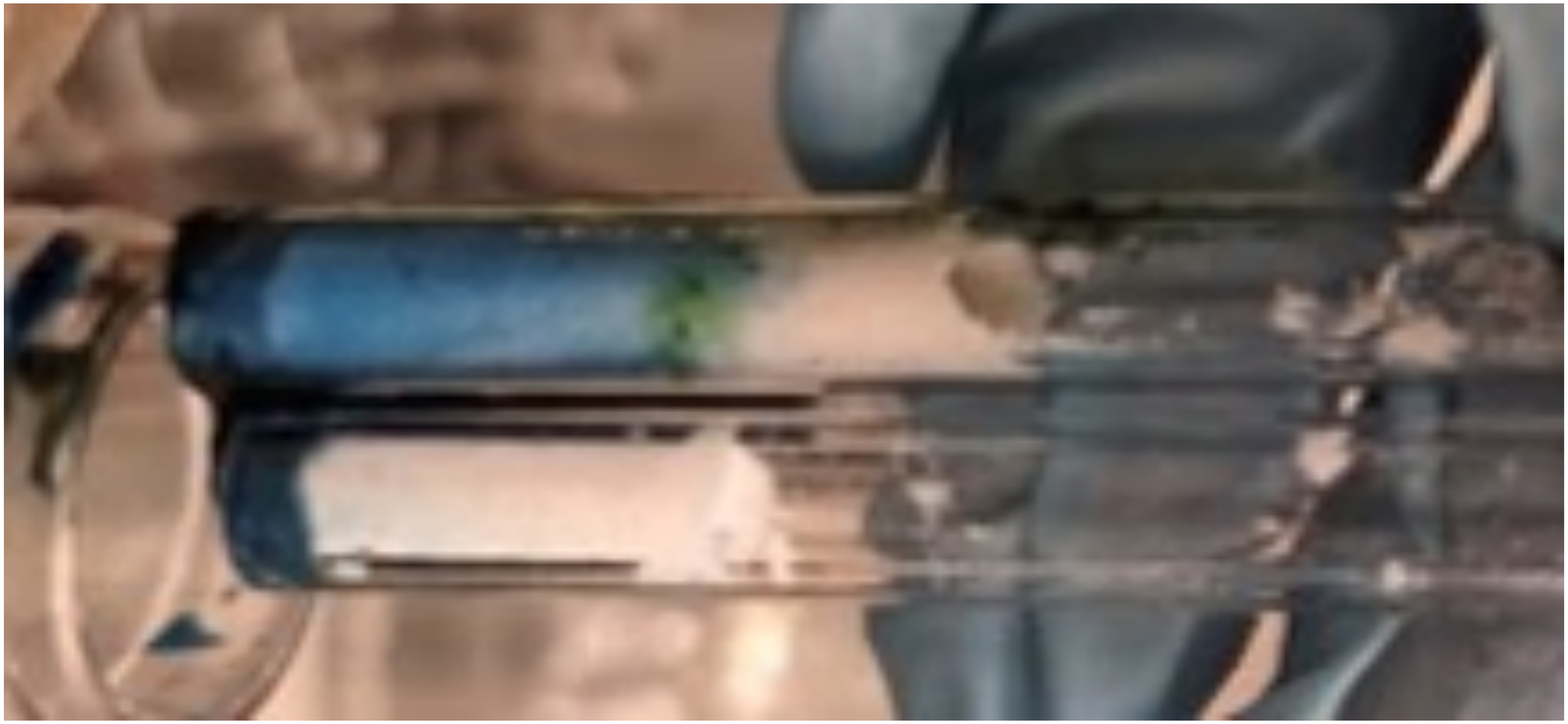

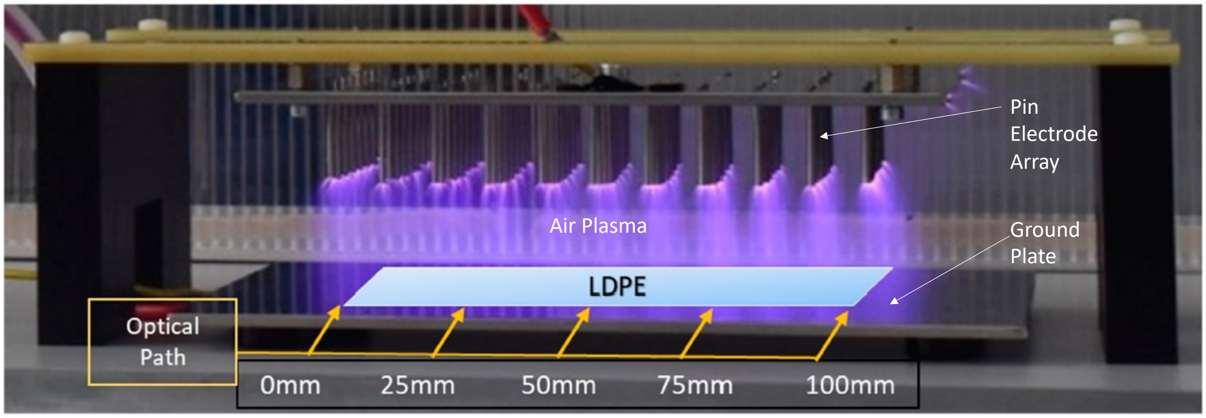
Pin
Electrode
Array
Air Plasma
Ground
Plate
LDPE
Optical
Path
0mm
25mm
50mm
75mm
100mm

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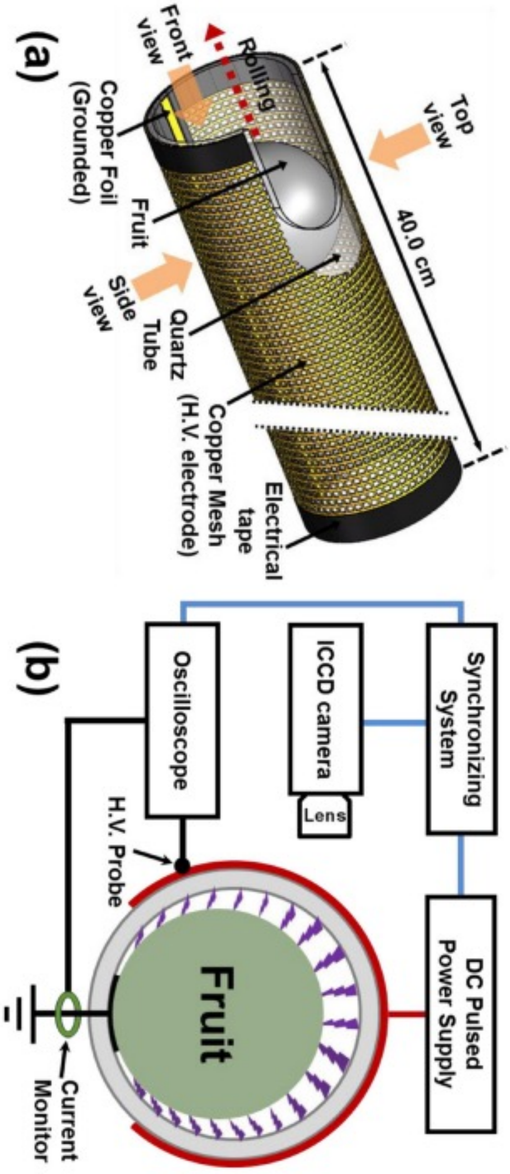

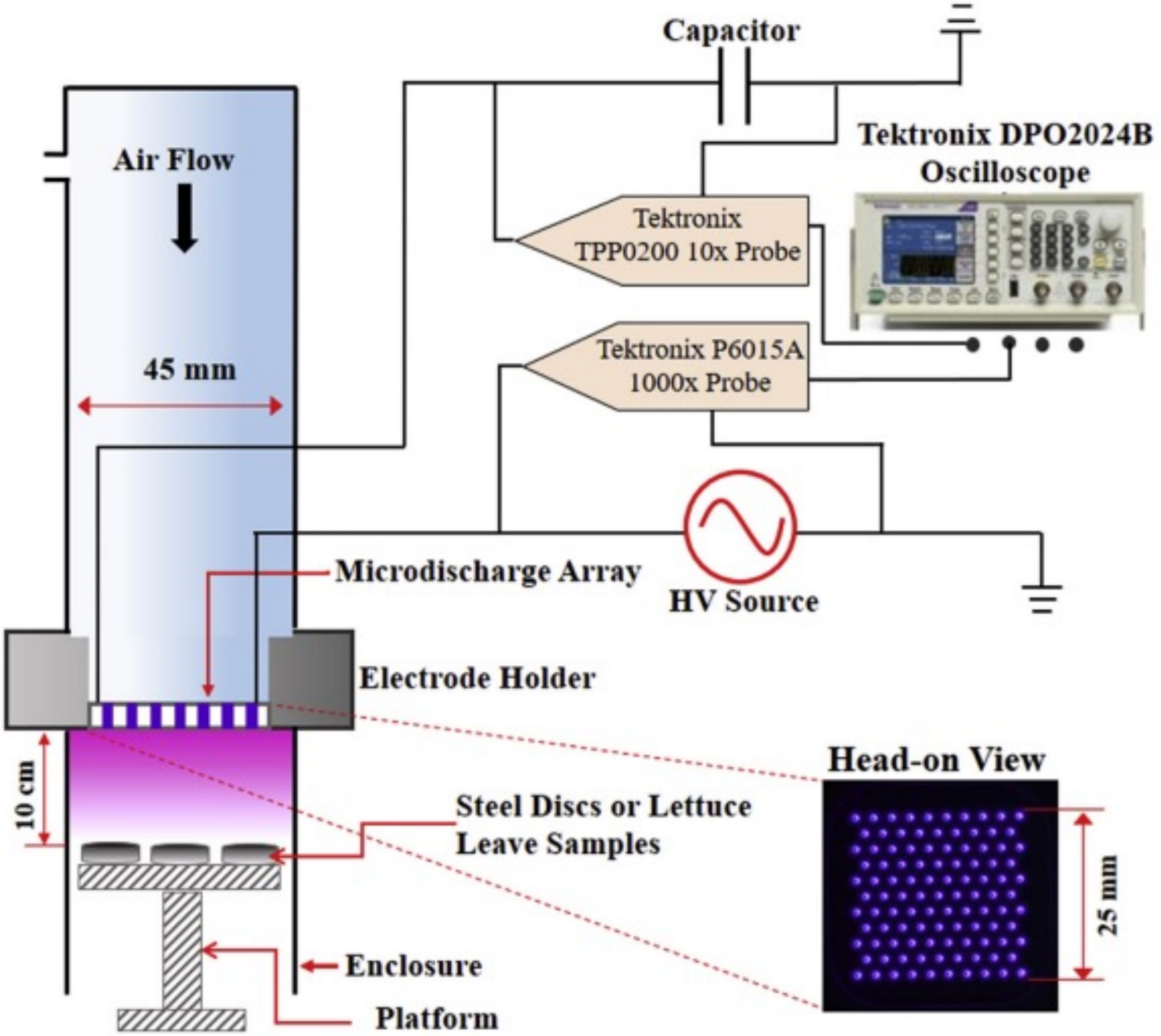
Capacitor
Tektronix DPO2024B
Oscilloscope
Air Flow
Tektronix
TPP0200 10x Probe
Tektronix P6015A
1000x Probe
45 mm
Microdischarge Array
HV Source
Electrode Holder
Head-on View
10 cm
Steel Discs or Lettuce
Leave Samples
25 mm
Enclosure
Platform

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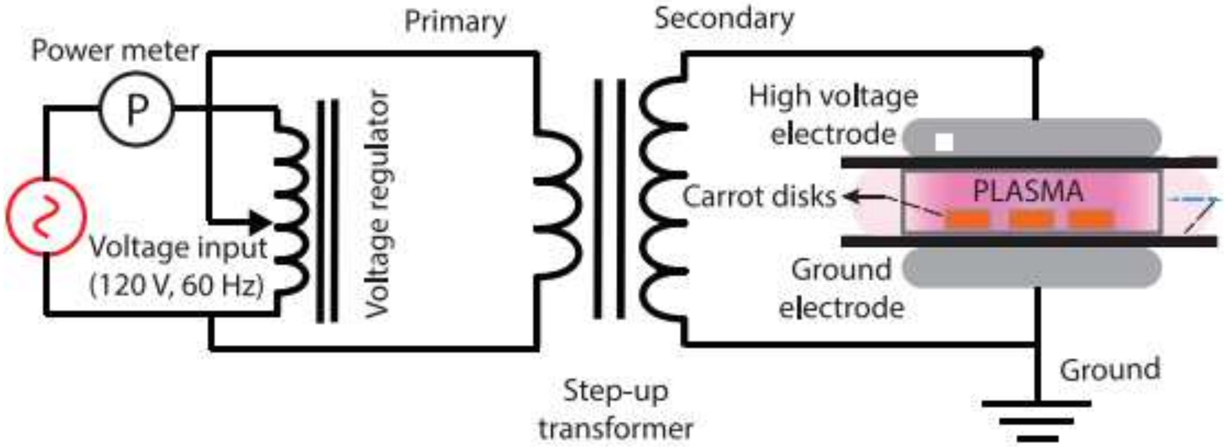

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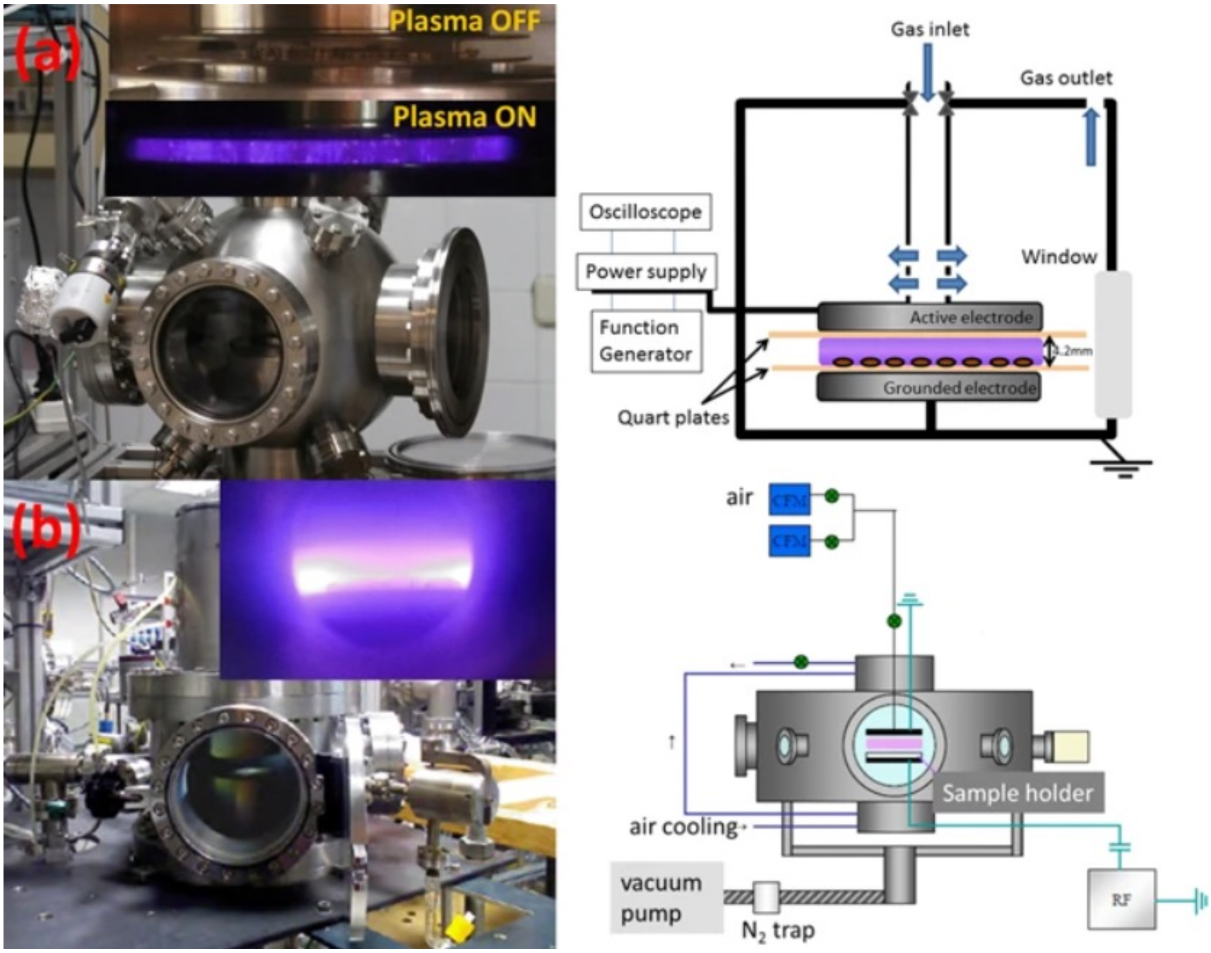

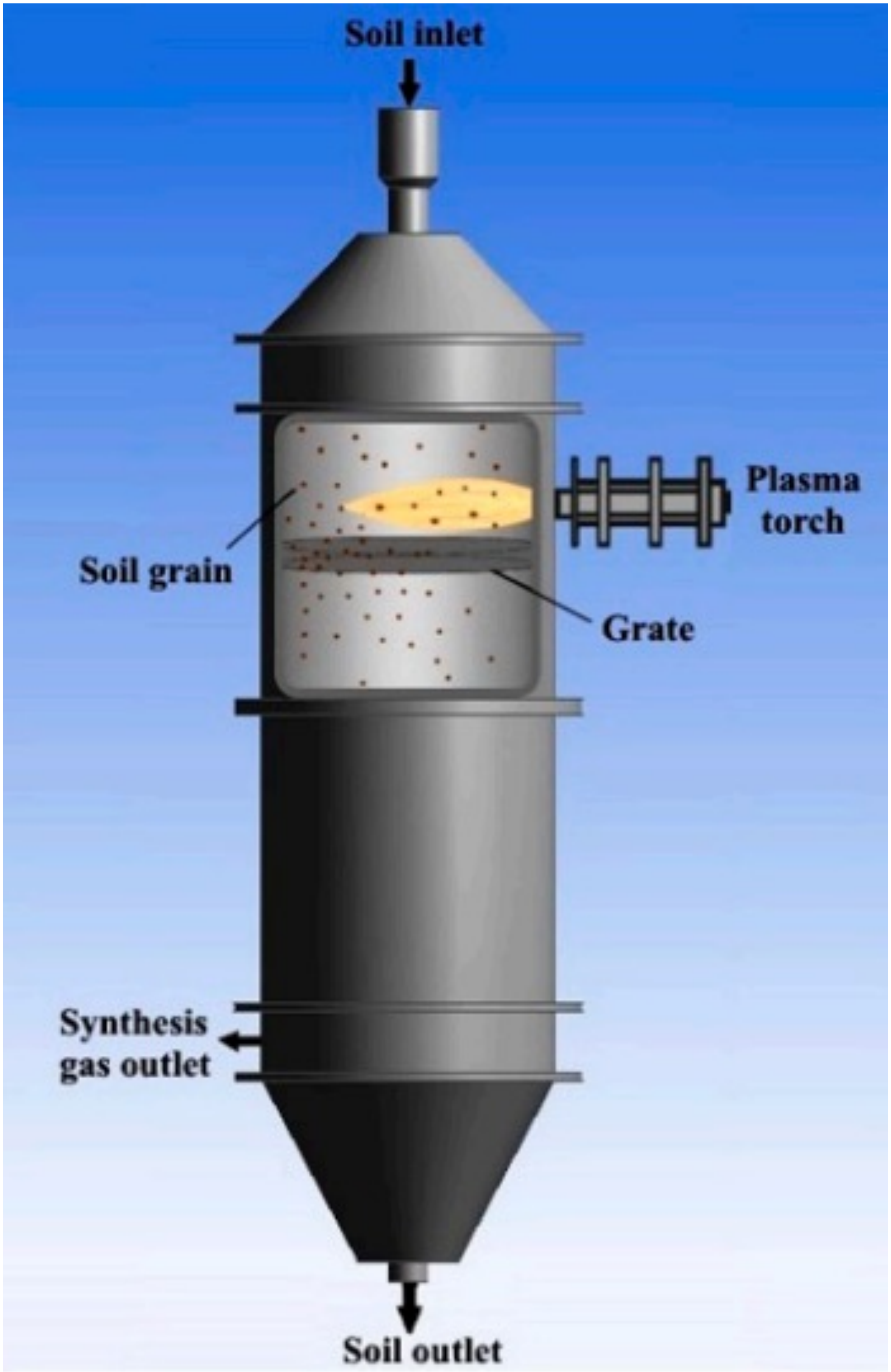
Soil inlet
Plasma torch
Soil grain
Grate
Synthesis gas outlet
Soil outlet