

# Mimicking Nuclear Radiation-Induced Chemistry with Friendly Cold Plasma

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## Abstract

Chemistry and related research that aims to address and solve myriad challenges is ultimately evaluated or judged from practical points of view. A successful endeavor generates a sense of satisfaction and opens new research directions; it may also substantiate the novelty and apt use of the experimental methodology chosen therein. Diversity in research strategies, aided by improvements and availability of related instrumental facilities, progressively allow more complex challenges, which seemed beyond reach previously, to be addressed and resolved. In this context, systematic estimations of any reaction's physicochemical parameters and its underlying stepwise mechanism in any occurrence have always remained an essential attribute of research. Such quantifications allow user control of chemistry if desired and help alter or enhance productivity selectively by tinkering with a 'reaction environment'. Chemical reactions' commencements almost always necessitate energy input in some form and often set out either in the oxidative or reductive milieu. **Nuclear Radiations (NR)** such as high energy  $\gamma$ - and X-rays (em radiation), and particles such as  $\alpha, \beta, H^+$  etc. from radioactive decay can create redox environments in a variety of media. Thus, for more than half a century, machines such as particle accelerators and gamma-chambers (in addition to non-nuclear sources such as LASERs and UV-vis light) were routinely deployed as sources for convenient and speedy evaluations of various types of reaction properties. Such measurements made the world over in different facilities and laboratories have played a pivotal role in enriching our shared knowledge base. In the last two decades, an exciting technology relating to **Cold Atmospheric Plasma or Cold Plasma (CP in short)**, with its inclusive reactive energy, has emerged and matured rapidly. It offers an undemanding alternative to the NR studies that stipulate the use of vastly expensive and very complex hardware systems. Though the history of CP science and technology spans over the last 150 years, it remained utterly dormant or sparsely researched until the 1980s, mainly due to

ignorance. At present, setting up a CP experimental facility or laboratory is an uncomplicated and generally an easy to finance proposition. Convenience in tuning CP generation currently makes it one of the user-friendliest technologies available for contribution in various chemistry-inclusive application areas, creating appropriate or innovative environments. Worldwide different laboratories, institutes, and even industries have found profitable use and application in diverse and utilitarian areas such as pollution control and mitigation, material surface modification, in biology, health and healing, nanomaterials and fabrications, polymers, improved vehicular engine performance, creating steps towards a cleaner and greener environment etc. to name a few. Herein, the "**tricks of the CP trade**" and some of its recent diverse multi-phase, atypical or innovative chemical and other applications, including some carried out in our laboratory, are presented to sensitize and motivate researchers about this novel topic for endless exploration.



**Tomi Nath Das** received his Bachelor's degree in Chemistry from the University of Delhi, a Master's from IIT Kanpur, and the Ph.D. from the University of Bombay, Mumbai. Starting 1978 till 2016, he worked as a Scientific Officer at the Bhabha Atomic Research Center, Department of Atomic Energy in various laboratories situated at Srinagar, Gulmarg and Mumbai. His research included homogeneous and heterogeneous atmospheric trace level chemical reactions, nuclear radiation and photo-induced free radical chemistry in condensed media, Cold Plasma induced gas-phase and on-surface chemistry. His Post-Doctoral Research at the National Institute of Standards and Technology, Gaithersburg, Maryland, USA, focused on the Mechanistic Free-radical Chemistry in Condensed Media. Since 2006 he initiated and led the Research and Development exploring Cold Plasma generations and their utilitarian Free-radical chemistry in multiphase environments and on living and non-living surfaces. Currently, he mentors Cold Plasma research initiatives in physical and biological environments at Ravenshaw University, Cuttack and KIIT University, Bhubaneswar.

## Introduction

Initiating a chemical reaction generally necessitates energy input in some form. Amid various sources available, the energy of **Nuclear Radiation** (NR for short) has a special place in the R&D as well as industry sectors, mainly due to its distinct character and properties. A brief account of NR application over the last seven decades is presented here to sensitize the reader about its established potentials and protocols followed during use. In the previous two decades, another exciting area of science and technology, namely **Cold Atmospheric Plasma** (CP), has emerged, imitating or mimicking NR applications. Moreover, CP in research and development is freely usable at a small fraction of the cost of NR and is also free from any associated intricacy of the latter. As a result, CP applications have rapidly gained acceptance worldwide for numerous purposes. For readers who are not yet familiar with this exciting area of research and development, an abridged account of CP generation and its functional roles in multidisciplinary environments is presented. The narrative aims to reveal the underlying CP chemistry duly controlled to assist its harmonious amalgamation in other domains, with select working examples that recommend its future innovative potentials in most spheres of human activity.

**Free radicals and the energy unit of electron Volt:** For the effortless perception of the following text, it is necessary to get unambiguously acquainted with two simple terms, namely **Free radical** and **electron Volt**. **Free radicals**, as defined and noted in the **IUPAC Gold Book**,<sup>1</sup> are atomic or molecular entities that have free or unpaired electron spin. These are represented (with superscript *dot*) as Atom<sup>•</sup> or Molecule<sup>•</sup> and may have an electric charge or be neutral. Typical common examples are a hydrogen atom, H<sup>•</sup>, mono-nitrogen oxides such as NO<sup>•</sup>, NO<sub>2</sub><sup>•</sup>, methyl radical, CH<sub>3</sub><sup>•</sup>, superoxide anion, O<sub>2</sub><sup>•-</sup> etc. Very familiar hydrated ferric ion Fe(H<sub>2</sub>O)<sub>6</sub><sup>3+</sup> would fall in this category and may be visualized as Fe(H<sub>2</sub>O)<sub>6</sub><sup>•3+</sup> though in general practice the superscripted <sup>•</sup> symbol is always omitted. Molecular oxygen needs a special mention here, it being a **biradical**, 'O<sub>2</sub><sup>•</sup>'. More importantly, though, the chemical reactivity of most **Free radicals**, especially the small sized ones, fall in the **ultrafast** regime (meaning almost every diffusion-controlled collision results in a product formation; therefore, such reaction time scales remain in nanoseconds). Some details of **Free radicals**' chemistry are presented later.

In chemistry, **Electron Volt** is a frequently discussed and adopted measure of energy, especially in the field of NR induced Chemistry, Photochemistry etc. One **Electron Volt** is the **kinetic energy** gained by a single unbound electron when accelerated by an electric potential gradient of 1 volt. The numerical value of one electron Volt (represented as **eV**) is  $1.602 \times 10^{-19}$  Joules (SI unit). Typically, UV photon of 310 nm wavelength energy is 4 eV. (For comparison, one 100 watt incandescent lamp uses

360000 Joules per hour or  $2.25 \times 10^{24}$  eV energy.) In the subsequent presentation, this unit of energy is used.

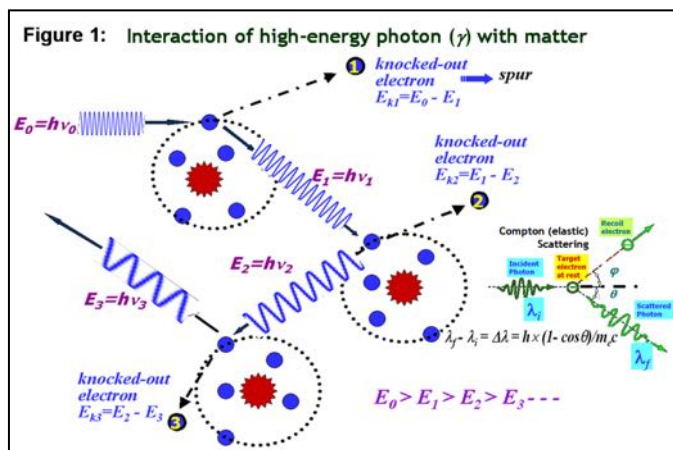
**Nuclear Radiation-Induced Chemistry:** A brief account of **Nuclear Radiation** (NR) here aims to relate its vast application in chemistry and industry and motivate the interested young readers and other researchers to explore this frontier area as a prospective career option, if desired (<http://www.barc.gov.in/careers/officers.html>). NR is synonymous with  $\alpha$ ,  $\beta$ , and  $\gamma$ -radiations emitted from radioactive nuclei and other charged particles (e.g.  $e^{-/+}$ ,  $p^+$ ,  $D^+$ ,  $He^{2+}$ ,  $M^{n+}$ ) from various processes and machines. Such sources include *natural radioisotopes*, e.g. <sup>3</sup>H, <sup>14</sup>C, <sup>40</sup>K, <sup>222</sup>Rn, <sup>all</sup>U etc. and Cosmic rays (including Solar origin); Man-made sources include Nuclear Power Plants and Related activities (mining, fuel processing, waste disposal), and Nuclear Explosive Devices. Typically, X-ray (discovered by W. C. Röntgen in 1895) photons with energy above ~125 eV to an upper limit of ~150,000 eV are also considered NR. Body & Dental X-ray Machines, other Medical Diagnostic Devices, Laboratory and Industrial NR emitters such as  $\gamma$ -Chamber and Particle Accelerators fall in the same category. Since NR are hazardous, all users and workers need to be aware of its presence nearby and follow stringent safety protocols. They are appropriately warned by the black and yellow **Trefoil symbol**.<sup>2</sup> This symbol was designed at the Berkeley Radiation Laboratory, University of California, in 1946 and subsequently adopted by the International Atomic Energy Agency (IAEA). New Supplementary Ionizing Radiation Warning Symbol is also available since 2007 for wider general recognition, even by the non-NR workers.<sup>2</sup> Users of X-rays, especially in the medical arena, are similarly warned by another appropriate symbol.<sup>3</sup>



For chemistry research and related activities in other non-research applications, the most preferred NR sources employ <sup>60</sup>Co isotope encased in stainless steel. Each radioactive <sup>60</sup>Co nucleus decays emitting two  $\gamma$ -photons of energy 1.17 and 1.33 MeV (or average photon energy 1.25 million eV, half-life 5.27 yr), allowing its use as a *steady* or *continuous* energy source. <sup>137</sup>Cs isotope source of sub-MeV NR ( $\beta$  0.512 MeV and  $\gamma$ -0.6617 MeV, half-life 30.1 yr) is also used for similar purpose.<sup>4,5</sup> In contrast, to investigate and characterize *reaction intermediates* having an *ultralow lifetime of survival* within any reaction environment (commonly mentioned as “*transient*” chemical studies), NR sources with matching *pulse width* such as Febetron, Van de Graff, Linear Electron Accelerator, and Synchrotron are being used for almost last seven decades.<sup>6-9</sup> The *transient* studies allow chronological quantification of reactants' and reaction's physicochemical properties and frequently also assist to indirectly “visualize” the progress of chemical reaction(s). NR induced chemistry research typically

encompasses gas, liquid, and solid media, as per user choice. It is befitting to note here that Prof. S R Mohanty performed his irradiation measurements on inorganic crystals employing a 2 MeV Van de Graff accelerator at Cambridge, Radiochemical Lab (+ve ion source of  $H^+$  and  $He^+$ , beam currents up to 100  $\mu A$ ) built by High Voltage Engineering Corporation, Burlington, Massachusetts, USA (See Author's Note<sup>†</sup> for a few selected publications of Prof. S. R. Mohanty).

The emission of  $e^-$  or  $\gamma$  entity in radioactive decay appears in femtosecond ( $10^{-15}$  s) timescale. In contrast, the charged particles from an accelerator mentioned above are beamed in timescales of pico-, nano-, or a few microseconds ( $10^{-12}$ ,  $10^{-9}$ ,  $10^{-6}$  s). The subsequent dissipation of NR energy in a target occurs stepwise and gets completed within the timescale of picoseconds ( $10^{-12}$  s). The primary energy loss mechanism that prevails at NR photon or particle energy of a few MeV (Table-1) follows the elastic **Compton Scattering** mode. **Figure 1** summarizes the scattering principle. The NR interacts with the electrons present in a target atom of a molecule. Higher target electron density or cross-section is offered by elements of higher atomic numbers. Similarly, higher physical density (solid >> liquid >> gas) of target material also allows a higher fraction of incident energy absorption.



**Figure 1** reveals that the loss of interacting photon or particle energy is gradual and in distinct steps, with a repeated generation of products even by a single NR entity. The knocked-out electrons from all stages re-interact and initiate chemical changes in spurs, which are distinct strings of discontinuous nano volumes with different chemical identities than the bulk target material. Additionally, the conjugate cation (**Free Radical**) formed due to electron loss may also initiate diverse chemistry.

In **Table 1** comparison of the numerical values of individual energy levels noticeably reveals why NR can initiate a multitude of similar *chemical identity change* steps within a

target of *atomic or molecular* nature. The extremely high energy content in each NR photon or particle causes multiple electronic excitations, bond dissociation, creation of structural defects in solids, ionization, etc. in the target during its interaction, including the formation of a variety of highly reactive **Free radicals** in the *ultrafast* timescale of nanoseconds, thus encompassing its worth in a variety of research.

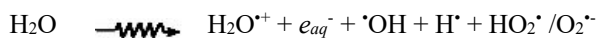
**Table 1. Typical Energies in Electron Volts**

gamma $\gamma$ -	0 – 3 MeV
beta $\beta$ -	0 – 3 MeV
alpha $\alpha$ -	2 – 10 MeV
<b>Diagnostic Medical X-ray photons</b>	<b>40-140 keV (0.14 MeV)</b>
Room temperature thermal energy of a molecule	~ 0.04 eV
Light photons: Visible / 100 nm UV	1.5 - 3.5 / 12.4 eV
Energy to dissociate $CH_3CH_2-H \rightarrow CH_3CH_2^{\bullet} + H^{\bullet}$	4.4 eV
Ionization energy of Helium	24.5 eV
Ionization energy of Argon	15.8 eV
Ionization energy of Hydrogen/Oxygen atom	13.6 eV each

In solid samples, NR use to create and study crystal defects are pretty common; other studies and applications include material purification and degradation, degassing, color generation (in diamonds), sterilization (medical devices and in food technology), polymers, surface modification (wood-plastics), determination of fossil age ( $^{14}C$  dating, *half-life* 5.73 yr), bio-solid waste treatment and purification or its value-added use etc. In some cases, low penetration depths of some NR particles are used to create changes at or close to the target surface, while with  $\gamma$ - or very high energy particles, more extended penetration within bulk material is effortlessly achieved.

In fluid samples and solutions, if the primary gas or solvent fraction is chosen high (such as dilute,  $\leq$  mM solute solutions in water/organic solvents), then within *nanoseconds*, NR energy gets homogeneously dissipated in it. After a radiation interaction event, a soup of reactive primary gas or solvent related **Free radicals** (ionic and neutral) and electronically excited states are created up to depths of mm to cm, depending on the target state, contents, and density. A variety of reactive initiators thus created produce environments with characters such as oxidizing, reducing, presence of ionic and neutral excited entities, etc. Simultaneously, with sophisticated and dedicated online detection and measurement instruments, a variety of consequent changes in the sample, e.g., spin density (in gaseous medium), optical characteristics (in gas and liquid), ionic density, pH etc.,

are often measured. These types of studies, when performed in real-time, also permit *ultrafast* stepwise mapping of physical changes and chemical reaction pathways that may lead to a variety of mechanistic and product generation results. Typically, in aromatic organic solvents, solvent triplet excited states that form first transfer energy to create solute triplets and help convenient evaluation of such properties (difficult to obtain otherwise by direct photochemical excitation studies). In aqueous solutions, the reaction after NR energy absorption occurs as follows:



The reaction symbol shown above is universally accepted to represent NR induced chemistry in a fluid medium. Similarly, in gaseous media (e.g. SF<sub>6</sub>), various types of NR induced chemistry have been studied. Due to low physical density, in this case, long path length sample cells are mandatory for sufficient and noticeable absorption of NR by the sample.

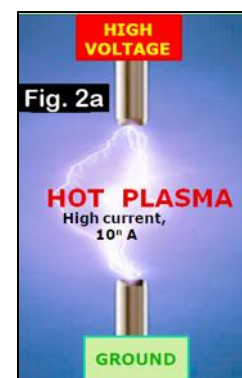
In summary, even in the most studied cases in liquid state and solutions, one finds a considerable and diverse role of NR. Some of its contributions therein include: (1) Characterization of physical, physicochemical, optical, structural, spin density, bonding, stability and reactivity, *redox* nature, and excited states' behavior of enormous number of **Free radicals**, ionic and neutral species; (2) Kinetic measures of related formation, lifetime, decay, and reactions' propensity in real-time under varying experimental conditions, e.g. in presence cosolutes and other reactants, *hν*, O<sub>2</sub> or other gases, added ionic salts, *etc.* with frequent mapping of reaction mechanisms; (3) Evaluation of thermodynamic properties like one-electron reduction potential, equilibrium constant, **Free Radical** acidity or pK<sub>a</sub> in varying medium pH; (4) Degradation or transformation to usable end products; (5) Studies related to biology and health, diet, antioxidation, drug actions *etc.* The list is truly endless. In the last seven decades, some areas where results have enriched our knowledge further include new and better methods in synthesis, creation of novel materials, innovative processes, alternate methodologies and technologies, cleaning of the environment, in medical science and biological processes, and applications in various types of industry. However, the major apprehensions in NR use are its extremely high cost to install a machine or facility; Large area or space required to house such facilities; Stringent requirements and application of safety norms for all users; and appropriate and sustained training to handle and use any type of NR source. Thus, NR facilities, though quite valued and spread over many countries, are limited in number and types, both for research and applications (though they remain quite prevalent in health and medical use).

In the context of NR utilization, a notable factor is it's close to a million-time higher *energy packet* compared to the actual necessity in a typical chemical step. Hence, *can another*

*energy source (mixed photon and particle type) with few electron Volt energy per entity possibly deliver a similar performance?* In the last two decades, **Cold Atmospheric Plasma** has emerged as the sure-shot and convenient answer. Mimicking NR, its contribution in diverse areas as a fundamental "**Chemistry Initiator**" developed from a straightforward generation strategy near atmospheric pressure with minimal complexity in related hardware. Its continually growing and widespread applicability makes it a practical choice for many. The remaining section of this presentation deals with the basics and relevance of CP and aims to encourage the reader to make an apt choice in future endeavors.

**Plasma at Atmospheric Pressure:** Plasma in physical sciences is known from 1928 when Langmuir borrowed the term from medical sciences (representing the fluid fraction of the blood) to describe a highly energetic state of matter, wherein the gaseous state transforms into a new form of ionized entities. Excessive energy gets pumped rapidly into the gaseous atoms or molecules, causing atoms and molecules to lose a fraction of bound electrons as kinetically energetic in a free or unbound state, with simultaneous formation of appropriate cations (mostly **Free radicals**). For example, ice/snow in the solid-state with energy input gets transformed into water in the liquid state. The latter give rise to its vapor with further energy input. Lastly, plasma species (*including a fraction in an excited state*) like *e*<sup>-</sup>, H<sub>2</sub><sup>•+</sup>, H<sub>2</sub><sup>•-</sup>, (H<sub>2</sub>O)<sub>n</sub><sup>•+</sup>, H<sup>•</sup>, <sup>•</sup>OH, O<sup>-</sup>, O<sub>2</sub><sup>•-</sup>, <sup>•</sup>O<sup>•</sup>, in addition to molecular species like H<sub>2</sub>, O<sub>2</sub>, O<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, (some unchanged H<sub>2</sub>O), and *hν* are produced from water vapor when the excessive amounts of energy are pumped into it under a variety of conditions. One of the methods for plasma creation within any gas *at or near atmospheric pressure* is via an "electric discharge". In a simplified description, the details are as follows.

It needs to be noted that the background cosmic rays and natural radioactivity *continuously* and *randomly* generate a very low concentration of ion pairs (freed *e*<sup>-</sup> and **Cat**<sup>+</sup>) in our surrounding atmosphere or any other gaseous medium. Typically, the steady-state concentration of such ion pair is < 10<sup>4</sup> m<sup>-3</sup>; as older ion pairs get neutralized or trapped and lost, simultaneously new ones are generated independently of the previous events. In this scenario, if appropriate magnitude potential difference (few kV mm<sup>-1</sup>) is applied between two separated points or surfaces inside a gaseous medium, as represented in **Figure 2a**, the freed *e*<sup>-</sup> being very light as compared to any **Cat**<sup>+</sup> generated, gets suitably accelerated towards the anode (+ve charged surface of electrode). It may gain sufficient energy to ionize more intervening atoms or molecules on collision during its flight,

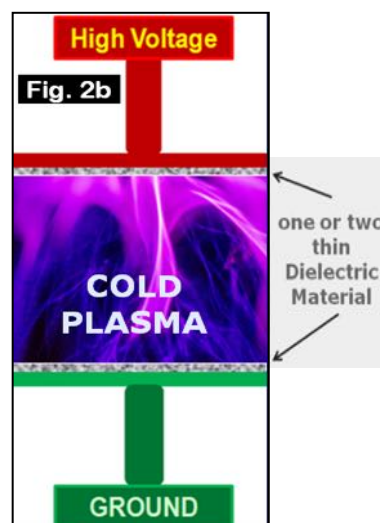


producing  $e^-$  multiplication or an avalanche (Townsend multiplication) within  $10^{-12}$  -  $10^{-9}$  s time scale. Under supportive conditions, the avalanches grow to create a random electric discharge. Parameters that play a crucial role include the *Prevailing Gas Pressure (near atmospheric pressure as discussed here)*, *Magnitude of Applied High Voltage*, *Inter-electrode gap*, *Electrode shape (curvature)*, *Nature of Gas etc.* Thus, continuous current flow (for time scale  $10^{-9}$  s and slower, and persist for the period the potential difference exists) may ensue between the two electrode surfaces. Subtle differences in the *electrical breakdown* of gases differ due to (i) Dissimilar electron-impact ionization processes in different atoms and molecules due to Internal Energy Partition dynamics; (ii) Target Atom Electronegativity, Ionization Potential *etc.* At atmospheric pressure lower than normal, fewer ionizations per unit volume happen due to longer *mean-free-path* at a lower density.

In contrast, at higher pressure, the *mean-free-path* gets shorter. Still, more frequent collisions cause a high fraction of kinetic energy loss in thermal processes (sufficient energy to cause successive ionization may not be gained by  $e^-$  before its premature collision). In effect, the higher breakdown voltage may be mandatory in either case. Typical breakdown voltage values *at atmospheric pressure* for various common gaseous media are He 1.56 (kV mm<sup>-1</sup>), Ar 1.73, N<sub>2</sub> 2.51, H<sub>2</sub> 2.73, Air 3.4, N<sub>2</sub>O 4.2, CO<sub>2</sub> 4.3, O<sub>2</sub> 4.5. In addition to the electron multiplication physical mode mentioned above, a fraction of the freed electrons (and a small fraction of cations) chemically interact with neutral or other ionic species present and produce electronic excited species, atomic/molecular clusters (both charged or neutral), **Free Radical** anions (by electron capture, mostly if strong electronegative atoms such as oxygen, fluorine are present), and may also cause fragmentation of some molecules (*typically organic*) present. Some of the electronic excited species created inside the ensemble decay in ultrafast time scale, emitting copious *em-radiation* covering the range of IR to vac-UV, always giving the plasma an intensely bright appearance. Well-known examples of such plasma are (a) lightning bolts we observe during a thunderstorm and rain, (b) a welding job, (c) an electric spark near a live wire, *etc.* Following medium breakdown as described above, an arc-like random electric discharge with current flow  $\sim 10^n$  A may prevail. The medium resistance breaks down completely, allowing free current flow as long as the potential difference sustains. Such uncontrolled and very high current flow within the plasma causes ionic density to rapidly approach a very high fraction. The plasma thermal condition inside reaches extremely high Kelvin (close to billion degrees in stellar medium, near million degrees inside lightning bolts, few thousand degrees in welding arcs), creating the well-known condition of **Hot Plasma**.

The average electron temperature,  $e_T$  ( $\gg 10^{3+}$  K) in Hot Plasma almost equals different ion temperatures,  $i_T$  (and also equals constituent gas temperature,  $gas_T$  and boundary or contact

surface temperature,  $cs_T$ ; the subscript  $T$  refers to the respective kinetic temperature, *e.g.* 1 eV value of  $e_T = 11604.5$  K (from the relation  $1.602 \times 10^{-19}$  J /  $1.381 \times 10^{-23}$  J/K, taking into account the Boltzman constant,  $k_B = 1.381 \times 10^{-23}$  J/K). Thus, Hot Plasma is frequently referred to as “**Equilibrium Plasma**” due to thermal equilibrium amongst its various constituents. However, unlike Physicists and Engineers, **Hot Plasma** is not a preferred direct energy source for researchers aiming for chemical and biological applications. Material identities may get lost, and fundamental particles like electrons, protons, *etc.*, may prevail. *Therefore, is it possible to tinker with and manipulate such an energetic environment and tone down to conditions conducive for Chemistry (and Biology)?*



**Friendly proffer by Cold Plasma:** In the context of high voltage driven electric discharge discussed in **Figure 2a**, it was beneficially observed that when one or two thin layer(s) of any **dielectric material** is/are interposed between the two metal electrodes, the resulting current flow, and consequently the electric discharge potency is significantly reduced to a mild level, and the effect emerges quite

conductive for carrying out chemical reactions. Standard dielectric materials include glass, quartz, mica, alumina *etc.* The applied high voltage now charges the exposed surface area of dielectric attached to the metal electrode to an *almost equal* magnitude. Its constituting molecules only get electrically polarized within the dielectric material, and free passage for electric current through its material gets barred. As a result, the overall circuit current flow gets restricted to the  $\mu$ A to mA range, and only very mild and controlled electric discharge occurs. As shown in **Figure 2b**, the plasma thus created remained “Cold”, and its characteristics reveal a very low extent of contents’ ionization ( $\leq 2\%$ ). The resulting **Cold Plasma (CP)** harbors various types of oxidizing and reducing atomic and molecular species due to many reactions within (described below in detail). It may be vital to note that various CP constituents do not attain thermal equilibrium, and only the electron temperature,  $e_T$  attains high levels ( $> 10^{3+}$  K). On the contrary, the  $i_T \approx gas_T \approx cs_T$  ( $\sim 10^2$  K) remain  $\ll e_T$ . Thus, CP is also known as **Non-equilibrium Plasma, Atmospheric Pressure Plasma, Dielectric Barrier Discharge Plasma etc.** (Although Cold Plasma generation is also possible employing very different technology of radiofrequency and microwaves, such setups need pertinent

generation facilities, frequently limiting their common or ready accessibility and applicability for interdisciplinary and in specific basic research use. Therefore, these plasma generation methodologies are not discussed here).

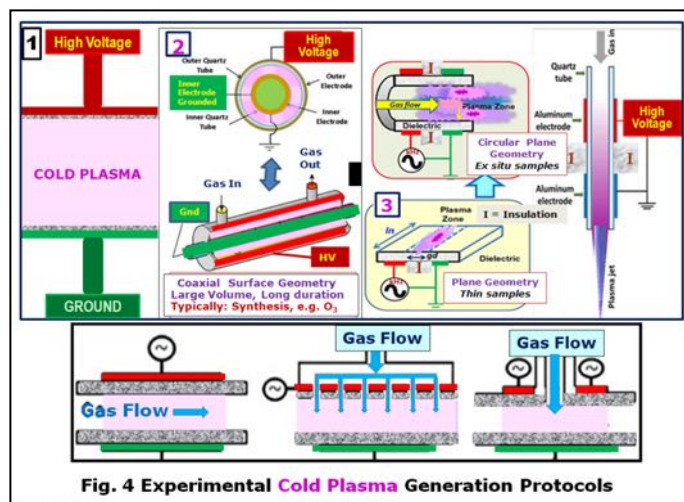
**Table 2.** Typical CP constituents produced in various gases near atmospheric pressure.

Gas →	CP constituents
He	$h\nu, e^-, He^*, He_2^*, He^{**}, (He)_n^{+/*}$
Ar	$h\nu, e^-, Ar^*, Ar_2^*, Ar_3^*, (Ar)_n^{+/*}$
N <sub>2</sub>	$h\nu, e^-, N^*, N_2^{*/**}, N_2^{+*}, N_2^{+*/**}, N_3^*$
O <sub>2</sub>	$h\nu, e^-, O, O^*, O_2^*, O_2, O_3$
Dry air 99% N <sub>2</sub> + O <sub>2</sub>	Primary = $h\nu, e^-, O, N^*, N_2^{*/**}/O_2^{*/**}, N_2^{+*}/O_2^{+*}, O_2^{+*/**}/N_2^{+*/**}$ Secondary = $O^-, O_2^-, N_xO_y, O_3$
Humid Air 99% N <sub>2</sub> + O <sub>2</sub> + H <sub>2</sub> O	Secondary = H <sub>2</sub> O <sub>2</sub> , HO <sub>2</sub> <sup>•</sup> , O <sub>2</sub> <sup>-</sup> , HN <sub>x</sub> O <sub>y</sub> etc. in addition to some of the above
CO <sub>2</sub> in moist air, 99% N <sub>2</sub> + O <sub>2</sub> + CO <sub>2</sub> + H <sub>2</sub> O	Secondary = C <sub>x</sub> O <sub>y</sub> , HCHO, C <sub>2</sub> N <sub>2</sub> etc. in addition to some of the above
Few % H <sub>2</sub> O <sub>v</sub> in > 90% He or Ar	Secondary = H <sup>•</sup> , H <sub>2</sub> , H <sub>2</sub> <sup>+</sup> , H <sub>2</sub> O <sup>+</sup> , •OH, O, O <sub>2</sub> , O <sub>3</sub> , O <sup>-</sup> , O <sub>2</sub> <sup>-</sup> in addition to primary He or Ar related some species as above

**Table 2** lists typical CP major constituents in various gases. Notice subtle differences related to the nature of species generated in the specific gas employed, mainly due to inherent differences in physical characters discussed previously. The characteristic wavelengths of the emitted light also differ in different gases due to the specific and dissimilar de-excitation pathways for various atoms, ions, and neutral entities created. The wavelengths measured by Optical Emission Spectroscopic (OES) thus reveal the light emitters' characteristics in any mixed soup and help identify the related species present within. Utilization of different gases and even mixtures of varying fractions can give rise to numerous possibilities regarding creating reaction conditions, either to mimic ones that prevail elsewhere or explore novel ones. Since He is an expensive gas, relatively cheaper and freely available Ar is frequently preferred in most studies.

Some randomly selected CP reactions and respective timescales of occurrence are shown in **Figure 3**. It may be noted here that various time events substantially match the time events observed in the case of NR use. Likewise, a very potent and reactive soup of **Free Radical** and non-radical species get created. Furthermore, near an open atmosphere, various primary CP constituents from another gas may chemically generate significant amounts of secondary oxygen and nitrogen (ROS and RNS) species. Therefore, in most cases, the subsequent

practical CP Chemistry falls within the domain of **Free Radical** induced and frequently ROS/RNS mediated changes. The "**Trick of the Trade**" consequently boils down to appropriate exploitation of the reaction parameters in any intended direction. However, before specific examples are presented, it is necessary to have a detailed look into the experimental CP generation tactics, precise design details, and typically associated hardware essential for successful endeavors. *It may be worthwhile to record here that high voltage mandates appropriate insulation of all exposed and conducting points and surfaces and the use of proper protection gears. Excellent earth line connection is part of the system design that must never be ignored.*



**Fig. 4** Experimental Cold Plasma Generation Protocols

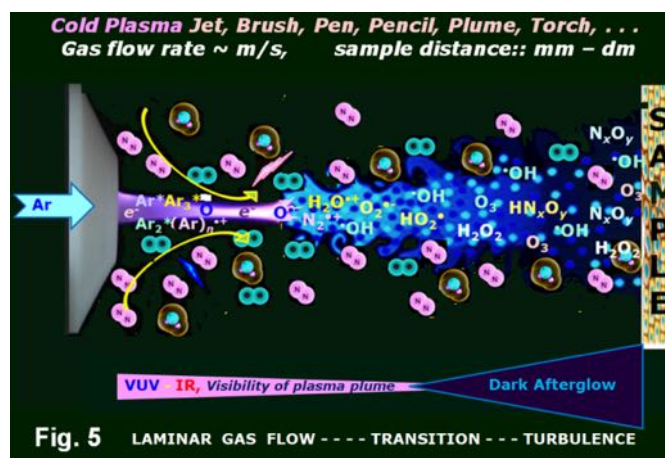
In **Figure 4**, some selected CP reactor designs are presented. **No. 1** is the most common parallel dielectric plate geometry discussed in **Figure-2b**. Note that the dielectric plates are thin, 1 mm or thicker. The dielectric plate plasma active surface area equals its shared opposite surface area with the metal electrode, attached contiguously without any gap on the backside. The linear gas-gap between the plate surfaces may be less than 1 mm to a few cm and necessitate the use of proportional high voltage for medium breakdown. Any sample, such as a flowing gas, liquid in a container, or solid sample, may be arranged or positioned inside this gap for direct exposure from CP contents. Design **No. 2** relates to a coaxial or cylindrical surface geometry of the dielectric plates. The reactant(s) in a flowing gas passes between the two dielectric surfaces (inner surface of the exterior metal-coated dielectric and the outer surface of the internal metal-coated dielectric). While liquid samples need protocols to handle, solid samples are conveniently accommodated within the gap. This generator geometry is typically used in ozone production (for almost the last one and a half-century, presented later). Design **No. 3** highlights the utilization of a single dielectric plate. In this plane geometry, two electrically separated and parallel strips of metal are coated or fixed on one side (the bottom as shown) of the

dielectric plate. The gap between the strips is adequately insulated. One of these strips is connected to the high voltage supply, and the other is grounded. Plasma generation on the intervening area on the top of the dielectric surface allows sample treatment. Furthermore, if the flat dielectric plate is imagined to be rolled into a tubular shape, the CP gets created and contained inside the tube volume. For treatment, samples may be placed within the active plasma zone. However, if the need arises, the active plasma may be carried out of the tube and into the open with subtle design modifications and by maintaining adequate gas velocity. Then sample exposure with the released CP jet (alternately called a pen, brush, plume, wand *etc.*) is possible at some distance from the active creation zone. This is currently a convenient and oft employed CP source design.

In **Figure 4**, various gas flow arrangements are shown, which are self-explanatory. In some cases, the complete CP generation system is housed in an enclosure for convenience or safety (especially if some obnoxious product needs to be isolated or handled separately or for isolating the system from an open atmosphere). Since high voltage is an integral part of the setup, appropriate supplies include DC, AC 50/60 Hz, Pulsed DC, High-frequency AC *etc.* types as per need or application. Each class offers different CP characteristics suitable for the design of specific setups. While many of these high voltage supplies are available commercially as compact table-top units, these are frequently designed and fabricated in-house if such technical expertise is available. During CP experiments in our laboratory, we sometimes found random *em-interference* from such devices onto other electronic measuring instruments or computers placed nearby. To overcome this severe difficulty, we designed and developed a portable, table-top CP Flexible Brush system (with its dedicated inbuilt power supply) wherein the primary CP was locally generated in a quartz tube, then it was “temporarily extinguished”, but with its “information of creation” was carried over a safe distance (6 - 10 feet) through a Teflon tube via a thin copper wire, and made to reignite as a secondary *ex-situ* CP plume on any sample, thus enhancing the device flexibility and its consequent adaptability.<sup>10a</sup> Similar complete basic ready-to-use systems may be fabricated or purchased for ₹50 – 100k. In this context, the reader may note that the familiar kitchen gas-lighter enfold a piezo-electric transducer inside, which on the application of pressure, produces a spark that ignites the cooking gas over a stove. The spark appears as the transducer generates a required level of high electric voltages (kV), sufficient to cause air+gas electrical breakdown. With appropriate modification, the same transducer has shown its ability for convenient CP generation in small local volumes,<sup>10b</sup> especially in many hand-held, immensely utilitarian devices now available commercially.<sup>10c</sup>

An example of Argon CP plume is presented in **Figure 5** to assist readers in visualizing its physical characteristics and

getting acquainted with the chemically transforming ensemble over its exposed length. As the narrow and laminar plasma zone exits into the open air from left to right (*e.g.* from a tube as discussed in **Figure 4, Part 3**), it carries primary mono-, di-, and trimeric Argon radical cationic, neutral, and excited species (with some  $e^-$ ) out of the tip. The surrounding air continuously gets sucked and mixed within the CP plume contents due to the high speed of Ar flow ( $\text{dm}$  to  $\text{m s}^{-1}$ , at  $\text{L min}^{-1}$  volumetric flow rate exiting through a narrow diameter). The CP plume contents get transformed (in  $\mu\text{s}$  –  $\text{ms}$  time scale) as chemical and physical interactions generate secondary oxygen, nitrogen, water-related atoms, non-radicals, and **Free Radicals** (neutral, cationic and anionic nature) partially in excited states.



With forward flow, the plume chemical and physical characteristics continue to change; its shape gets into a turbulent mode and widens considerably. Diminishing plume visibility also suggests that the fraction of light-emitting (*electronically excited molecular and atomic*) species survive only a few milliseconds outside the tube. In experiments with large gaps, light emission dies down within a certain distance from the exit tip, and a dark afterglow region results. The chemical nature of the constituents continues to change further as de-excited and uncharged atomic and molecular species and other **Free radicals** get created. Finally, some of these “chemical contents” reach any sample placed downstream. Thus, it is quite apparent that factors such as the primary gas flow rate, plasma tube exit diameter, distance of the sample from tube exit, surrounding air composition *etc.* allow the user to design and carry out experiments to employ diverse primary, secondary, or even tertiary chemical initiators of varying reactivity. It may also be noted that replacing the surrounding air (using a suitable enclosure) with a chemically different environment help to change the nature and type of secondary species. For example, adding a monomer vapor in argon matrix allow controlled and an exclusive polymer coating over a sample surface. The user may even plan sequential or mixed layers for creating innovative alterations, making this simple arrangement throw up novel

ideas and applications. Even avoiding all extraneous chemicals in a closed chamber may conveniently achieve progressive surface cleaning, otherwise controlled etching, chemical modification, append alternate *redox* effects, *etc.* Lastly, intense vac-UV and visible radiation of energy 1.5 - 10 eV in the **CP** also allow users to exploit this energy for myriad applications. Thus, the possibilities are truly endless. Following randomly selected studies emphasize the versatility and practical potential of **Cold Plasma**.

**A. CP in Biology and Health:** A purist may suggest that molecular-level changes in biological contexts are relatable to basic chemical and physical steps. Thus, one may also wonder if **CP Physics & Chemistry** has relevance in any biology domain. The classic book “**Plasma Medicine**” by *Fridman and Friedman* published in 2013 (Wiley) amply provides wide-ranging answers, all in the affirmative. A review suggests that continuing basic research remains essential to improve, optimize, and enlarge the spectrum of safe medical applications of **CP** and monitor and control plasma treatment and its effects.<sup>11</sup>

Humans face seven most common biological enemies, which are sources of variable trouble. These include **viruses, bacteria, mold, pollen, dust mites, pet dander, and various pollutants**. We are already aware that **CP** can continuously provide **ROS, RNS** and other types of **Free Radicals** and intense UV-vis light at the flick of a switch. In this scenario, some recent examples below are intended to motivate readers to explore further, seek answers, realize the true potential of **CP**, or even contribute actively to find new cures in this ever-changing world.

Currently, the most discussed virus, SARS-CoV-2, in its various avatars, has put the world in a topsy-turvy mode. Thus, the leading inquiry is for a suitable **CP** counteraction to tackle this situation. The answer is YES, and seemingly quite potent! For example, the UCLA group has recently demonstrated that the virus is killed with **30 seconds** of **CP** assisted disinfection treatments on various surfaces, **including plastic, metal, cardboard, cotton from facemasks**, and even rough and wrinkled **leather surfaces** on basketball, football and baseball.<sup>12</sup> Similarly, the Max-Planck Soc. in Germany, in collaboration with Terra-Plasma Medical, has modified a previously developed hand-held device for chronic wound treatment into a direct application **CP** mechanical ventilator for Covid patients.<sup>13</sup> On another front, the next generation of protective masks are under development at the University of Michigan, which aims to harness the power of **CP** into a small **headset that both blocks and neutralizes airborne pathogens** (including virus), thus allowing free movement of the wearer.<sup>14</sup> Researchers at the University of South Australia have demonstrated that an “**Enhanced Cold Plasma protocol**” that otherwise helps eradicate antimicrobial-resistant bacterial infections in diabetic foot ulcers **also significantly reduces SARS-CoV-2 viral load**.<sup>15</sup>

Their strategy to utilize **CP** based “on-demand” amplified antimicrobial formulation comprises a rich mixture of highly oxidizing molecules: peracetic acid, hydrogen peroxide, and other reactive species such as Tetraacetythylenediamine and Pentaacetateglucose as antimicrobial precursor acetyl donors, which amplify the antimicrobial effects of **CP**. Thus, the targeted virus and a host of other bio-active enemies can be quickly and effectively neutralized with innovative onsite production and utilization of **CP**. With such success, **CP** has opened up a vast arena for research on its all-round antimicrobial potency and more future technologies for exploitation.

For many decades, the major health concern has been Cancer in its various forms. In this context, **NR** from either <sup>60</sup>Co radiotherapy machines, *e.g.* “**Bhabhatron**” developed in our country, or dedicated accelerator based devices are sources for targeted treatments. The use of specific radioisotopes is well known. Therefore, it is pertinent to inquire if **CP** can mimic and contribute likewise in a complementary mode. On an affirmative note, more than a decade ago, in 2011, pioneers in this *field* revealed the “**possibility of a paradigm shift in cancer therapy**” with **CP**.<sup>16</sup> Research carried out then found enormous potential to transform and enhance the available treatment technologies by deploying dedicated **CP** devices. After that, in 2013, the Max-Planck Institute for Extraterrestrial Physics in Germany reported the synergistic effect of **CP** with Chemotherapy on aggressive **brain tumor (glioblastoma) cells**, hailing the combination treatment as a new hope.<sup>17</sup> (*The same institute has also developed several table-top and hand-held CP devices to treat and cure Cuts, Wounds, Nail fungus, Ulcers, Bacterial and Fungal Infections*<sup>17</sup>). In 2015 it was revealed that the assortment of **ROS** and **RNS** type **Free radicals** were readily participating as potent weapons against Cancer, thus emphasizing their role and importance of appropriate physical exploitation of chosen chemical processes at nano-micro levels on site.<sup>18</sup> In 2019, an exciting and long-anticipated development by Purdue University thrilled the world, probably ending any scepticism and waiting.<sup>19</sup> It revealed a “**New CP apparatus to Kill Cancer Cells in the Colon**”. The US FDA approved the “**Canady Helois CP Scalpel™**” to remove microscopic cancer tumors remaining from surgery for first-ever use in a clinical trial. Further research in this direction continues.<sup>20</sup> It is hoped that the above account would motivate readers to explore further, participate and exploit, to achieve more such excitements with **CP** applications.

Hand sanitization activity before 2020 was habitually limited to frontline medical workers and some others in specific jobs. It was mostly ignored by the common populace worldwide. However, this activity has held newfound importance and necessity in the last two years. Previously, typical hand sanitizers contained chosen chemicals like **Triclosan, Benzalkonium Chloride, Providone Iodine** *etc.* However, it is not unknown that these non-alcoholic concoctions are not entirely friendly in the long-time application and use and are



linked to problems like dermal irritation, allergy, and toxicity of various types. With the advent of and late preference for alcoholic sanitizers, especially in the last two years, other types of warnings have simultaneously appeared from the FDA and researchers.<sup>21</sup> These include headaches, nausea, and dizziness after frequent use, especially in closed environs. Suggestions include soap and water being superior to sanitizer; preferably **hand washing if available** needs to be followed, as was a common past practice. In this context, however, it is pretty revealing that **CP-based** hand dryer cum sanitizer has been commercially available since 2013, with the claim of a 93 - 99% kill rate against a variety of pathogens tested within a short time.<sup>22</sup> Some of the other commercially available products for similar usage are presented later. An immense potential to research and develop standalone or portable commercial devices for common or targeted use awaits the interested researchers in S&T.

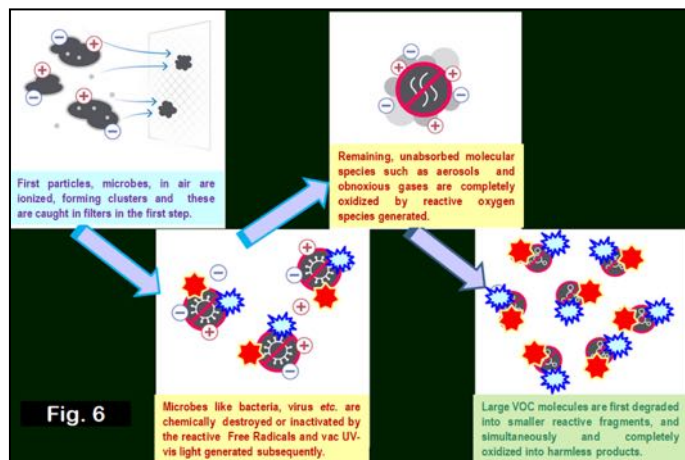
Within the last few decades, mobile phones and allied devices have become our inseparable friends. Following a wide variety of user activities, these are constantly exposed to biological entities released in user proximity and close contact with other surfaces, *etc.*, as the user travels and rests. Even the presence of faecal contaminants is not unknown. No wonder it is believed that “*The mobile phone not only remembers telephone numbers but also lack of personal hygiene and types and history of physical contacts*”. More recently, it is believed to be a forgotten or ignored *carrier source* of the SARS-CoV-2 virus.<sup>23</sup> **CP**, however, has proved to be reasonably practical in this regard, and back in 2015, an efficient “mobile sanitizer” was successfully researched, which revealed a 10 min treatment with very potent  $\cdot\text{OH}$  radical laden **CP** environ was sufficient, without affecting regular phone operation in any manner.<sup>24</sup> Further efforts in future are expected to turn out pocket-friendly units, *ca.* piezo-electric generation based or like a mobile case with **CP** generation as per specific user need.

Other related developments include creating **CP** devices and machines for applications in Dentistry to carry out Restoration, Treat Oral Infections, Teeth Whitening *etc.*<sup>25</sup> The active Free Radicals and other species in such use are reported to be  $\cdot\text{OH}$ ,  $^1\text{O}_2$ ,  $\text{O}$ ,  $\text{O}_2^-$ ,  $\text{NO}$ . These are currently awaiting FDA clearance. In the author’s laboratory, a simple Ar gas-based hand-held **CP** Electroporator was developed in 2015 to introduce extraneous matter (DNA or drugs) into the cell through partially opened cell walls, followed by its self-repair. It was observed that the same arrangement could be profitably employed to kill *E coli* if so desired. Thus, inactivation of microorganisms within a short time by non-reversible chemical destruction of cellular macro structures and molecules, including DNA, has been the “*trade-trick*” in Sterilization, Treatment and Healing of infected and chronic wounds, ulcers and burns, Increased coagulation and sterilization of blood, Biocompatibilization of surfaces, Aesthetic and ophthalmology

applications, Selective inactivation of cancer cells and healthy cell regeneration. It is vital to note that, unlike **NR** treatment, the patient is never exposed to any radiation. Additionally, during **CP** treatment, electricity does not flow through the skin or body. The operators of such devices or machines also do not face any risk for health. Other hallmarks of simple and controlled **CP** usage are low installation and process management costs, markedly reduced treatment times, and the absence of chemicals. The R&D scenario currently reveals a healthy effort in all-round development in this area.

**B. CP in Chemistry Research:** Few random examples highlight and encourage researchers who choose to remain engaged in the pure Chemistry domain to explore possibilities to accept **CP** in their daily work. (a) Production of 10 - 100 nm-sized *iron nanoparticles* from ferrocene in an alcohol solution at minimal cost and effort was achieved in a reductive environment offered by pure Ar **CP** jet for potential applications in water purification special magnetic micro- and nano-electronics usage.<sup>26</sup> (b) Graphene-supported *palladium nano-catalyst* (~ 50 nm) was conveniently made with **CP** for hydrodesulfurization of carbonyl sulfide (COS) in coal gas.<sup>27</sup> Almost 25 times enhanced catalytic activity was observed compared to Pd supported on activated carbon or Graphene-supported palladium composite prepared by the conventional reduction process. (c) Creation of High-performance *Platinum nanoparticle* embedded  $\text{TiO}_2$  photocatalyst for dye degradation.<sup>28</sup> The process involves simple steps like mixing  $\text{H}_2\text{PtCl}_6$  solution with nano  $\text{TiO}_2$  suspension for 24 h, followed by drying at 100 °C for 2h, and sample treatment in reducing **CP** of Ar and  $\text{H}_2$  mixture. The TEM of 1.5 wt% Pt/ $\text{TiO}_2$  suggests the formation of ~ 100 nm-sized catalyst and almost 25 times enhancement in its activity. (d) First time report from author’s lab of an efficient and straightforward *one-step conversion* of a few thousand ppm aromatic *benzene* to exclusive *aliphatic compounds* such as (i) methane (without catalyst, and enhanced and exclusive generation with added  $\text{H}_2$ ) or (ii) ethylene (with packed  $\text{TiO}_2$  coated catalyst within the **CP** generator, type-2 in Figure 4), both in moderately flowing Ar gas.<sup>29</sup> (e) Online transformation of abundantly available *C1 to C4 light hydrocarbons* (mainly methane) into usable liquid fossil fuel (*petrol or gasoline*, C5 to C12, and *diesel*, C12 to C20) was achieved in a single step with **CP** in combination with **Solid Oxide Electrochemical Cell**.<sup>30</sup> The latter assisted in minimizing radical recombination reactions, eliminating the formation of undesirable products. The other byproduct was water (no CO or  $\text{CO}_2$  formed). A web search for similar exploitable and novel **CP** use and applications in the field of Chemical science and technology reveals a rapidly exploding number of studies, with significant simplification of reaction designs with a reduced number of steps, dip in energy usage, avoidance of unnecessary chemicals and solvents in specific cases as is preferred or desirable for a “green technology”.

**C. CP for Environment Improvement:** Presentations so far suggest that the CP approach can contribute significantly in cleaning up our environments, which have been irrationally and uncontrollably allowed to degrade with time. In this section, the current scenario is reviewed.



**Figure 6** summarizes the various reactive cleaning steps and processes offered by CP presence. All unacceptable entities are taken care of by one or more of the four distinct steps, not necessarily in particular order or sequence. Random studies, processes and products are summarized below for a closer look at the level of development over the years.

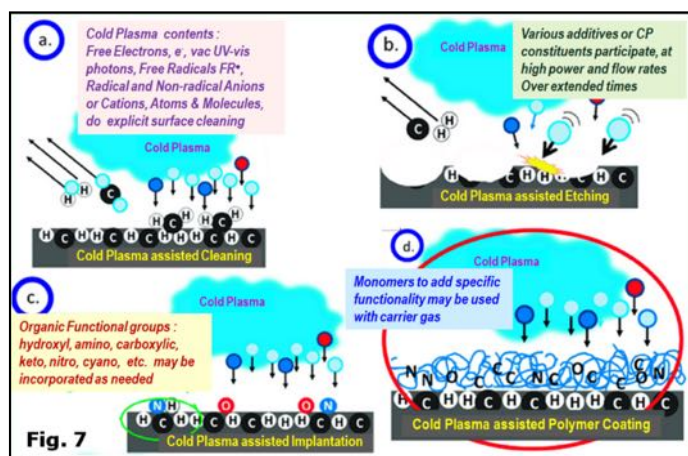
(a) **Indoor CP deployments:** In a recent study, inactivation of indoor airborne viruses is reported with the use of a glass bead packed bed CP reactor (type-1 in **Figure 4**).<sup>31</sup> A disposable filter near the exit removes all leftover particulate material. At a high airflow rate of 270 Lpm, the device continuously purifies air by 3 orders of magnitude. Imagine such devices placed in your laboratory at strategic locations; mountain fresh air may be available to you with just the flick of a switch! Of course, more than one commercial product is already available in the international market, including India, for suitable purification of indoor environments; these also include small portable ones for travellers for use in cars as well as hotel rooms, to a larger model for fixed use in larger homes, rooms, schools, hospitals *etc.*<sup>32</sup>

(b) Typical examples for **outdoor CP deployment** (very large capacity systems) presented below were researched and developed for specific purposes. (i) A 510 MW<sub>elec</sub> coal-fired thermal power plant in the USA is reported to utilize a three-stage process that includes a CP reactor, followed by a combination ammonia scrubber and absorber unit to neutralize and absorb the various acids formed; and finally, a wet electrostatic precipitator to remove the mists and aerosols.<sup>33</sup> Consequent reduction achieved for various species are as follows: SO<sub>2</sub> - 98 %, NO<sub>x</sub> - 90 %, total Particulate - 99.9 %, Fine Particulate - 95 %; Mercury - 80 -90 %. (ii) A patented Gas-

Phase Cold Plasma (GPCP) reactor was designed and developed at the Pacific National Laboratory, the USA, as a cost-effective and environmentally favorable alternative to the prevailing thermal removal of lethal pollutants and volatile organic contaminants (VOCs) from gas streams.<sup>34</sup> Reported destruction efficiency therein of various hazardous air pollutants as well as *chemical and biological warfare agents* are as follows: Trichloroethylene > 99.9%; Perchloroethylene > 99.9%; Benzene 97.85%; Naphthalene > 99.9%; GD nerve agent > 99.8%; Hydrogen cyanide > 99.4%; Cyanogen > 99.8%; Methyl cyanide 98%; Phosgene > 99.84%; Methane > 97%; Freon14 ~ 70%; Phosphonofluoric acid > 99.8%; and Dimethyl methyl phosphonate > 99%.

(c) **Potable or safe and clean water** is already a luxury at various locations and countries on our blue planet earth, mainly due to rampant exploitation of pristine natural resources and additional pollution from numerable sources. CP based solution in this direction is constantly getting modernized. Chlorine was the gas of choice for water treatment in the distant past. However, it was realized that Cl<sub>2</sub> gas is less potent for microorganisms, useless against chemical contaminants, and in the presence of certain polluting chemicals, may form carcinogenic trichloromethanes and chlorinated phenols. Thus, its deployment was eventually replaced by ozone, O<sub>3</sub> generated on-site from air or oxygen. The reactor design **Figure 4**, no 2 has been the mainstay for CP based O<sub>3</sub> generation, a safe, cheap, and one-step process controlled by the flick of a switch and needs only minimal human intervention in the form of maintenance. No wonder the world over the current estimate of O<sub>3</sub> production for water purification may be higher than 10<sup>12</sup> ton yr<sup>-1</sup> (~ 32 × 10<sup>3</sup> ton s<sup>-1</sup>). Ozone destroys bacteria, viruses, spores, mold, and algae also oxidizes most harmful chemicals such as dyes, insecticides *etc.* It is well known that excess O<sub>3</sub> quickly decomposes into O<sub>2</sub> in air and as H<sub>2</sub>O<sub>2</sub> in water, and finally into H<sub>2</sub>O and O<sub>2</sub>. Various small and portable table-top O<sub>3</sub> generators are commercially available for vegetable fruit cleaning at home and elsewhere. In such cases, the dissolved O<sub>3</sub> or its daughter product in water, mainly the **Free Radical** ·OH, efficiently completes the intended clean-up job. More recently, however, it has been reported that stubbornly resilient and toxic poly- and perfluoroalkyl (PFAS) compounds, sometimes found in contaminated drinking water (*from overuse of Teflon type coatings on kitchenware followed by its slow degradation*), are not decomposed and removed by O<sub>3</sub>. An innovative CP technology has been successfully researched for the hassle-free and successful removal of such chemicals.<sup>35</sup> It utilizes a **semi-hot Plasma**, first created is flowing air, directly injected into the contaminated water inside a reaction chamber. The suitably transformed CP with its “*afterglow reactive chemicals*” (Refer to **Figure 5**) destroy all the *otherwise impossible to destroy species* present. Very recently, a commercial water purification product has appeared in the market that claims a paradigm shift in “*oxidative water treatment*”.<sup>36</sup>

Thus, **Cold Plasma** S&T allows Environmental Remediation at low cost, with onsite usability, and is capable of running on solar energy, with minimal equipment complexity, and immensely exploitable and locally modifiable for chemical degradation and total removal of pollutants such as *VOCs* (volatile organic compounds including carcinogens, hazardous gases, harmful coloring agents and chemicals, bacteria and allergens (in humid and aquatic waste), particulates (such as aerosols), obnoxious odor (emanating due to presence of aldehydes, ketones *etc.*, H<sub>2</sub>S (sewage or industrial), NO<sub>x</sub> and SO<sub>x</sub> (fossil fuels' burning and other industrial sources), and dust mites frequently found indoors.



#### D. CP in Material Surface Changes and Improvements:

**Cold Plasma** being a surface-active contrivance, its convenient exploitation of solid samples and materials as per specific requirements has been widespread. In **Figure 7**, various modes of **CP** assisted surface treatment are shown, which are self-descriptive. Some random studies covering a wide range of aims and objectives are discussed below. In the domain of "New or extra *functionalization* of **Textiles**' Surface(s)", various materials were treated to add explicit surface properties. For example, wool, nylon, polyester fabric surfaces were altered to *hydrophobic* from *hydrophilic* with precursors such as styrene, dodecyl acrylate, and butadiene added in **CP** created in different gases.<sup>37</sup> Similarly, properties like total hydrophobicity, color-latching, superior appearance and feel with crease control, fire resistance, controlled porosity, antimicrobial character *etc.*, were incorporated by treating fabrics with **CP** in air, O<sub>2</sub>, Ar, F<sub>2</sub>, He, CO<sub>2</sub> or mixtures. Quite interestingly, the **CP** technology could be employed to manufacture "dual functionality" fabric, *hydrophobic* on one side and *hydrophilic* on the reverse.<sup>38</sup> Creation of *self-cleaning* cloth may get a boost with **CP** technology.

**Wood**, having a complex organic nature, is similarly amenable for surface chemical modifications. It is no wonder that the **CP** S&T has been extensively used to treat and

chemically modify wood and derived timber surfaces.<sup>39</sup> Superior finish and facile adhesion of surface polymer composite layer (wood-plastic combo) were thus achieved. This treatment method minimizes fungi growth and termite attacks and also helps to incorporate surface water and scratch resistance characters. A clean wood sample, e.g. a finished bathroom door, moving on rollers, is sprayed with a chosen liquid monomer and immediately exposed at 360° geometry to **CP**. The results are instantaneous and long-lasting. (Similar results had also been achieved employing **NR**; thus once again, **CP** indeed acted as a mimic, but comparatively at the token strain on resources and pocket)

Similar chemical creations of new and exotic surfaces include a few examples below. In a study to create photocatalytic, self-cleaning, and anti-staining *Super-hydrophobic* surface, the desired *nanoparticle + nanocomposite* combo surface coating could be effortlessly achieved using a dispersed aerosol of oleate-capped ZnO in n-C<sub>8</sub>H<sub>18</sub> in flowing He gas **CP**.<sup>40</sup> The coating combined the chemical characteristics of both the NPs and the organic component, which originated from the plasma polymerization of *n*-octane. In another study, **CP** assisted liquid deposition of -CF<sub>2</sub>-CF<sub>2</sub>- group on various substrates such as cotton, polyester, polypropylene, nylon, etc., using liquid aerosols helped create an oil, water, and alcohol repellent (~100%) surface coat.<sup>41</sup> Similarly, a **CP** assisted powder deposition technology has been patented that employs **CP** in combination with dispersed nanopowder to create a nanoporous nano-structured ceramic surface coating of 10 nm – 10 μm thickness.<sup>42</sup> In the author's laboratory, while developing a simplified surface cleaning process for Silicon Surface Barrier Detector before detector fabrication (*for later use in NR detection*), use of 90% Argon and 10% air-based **CP** for 3 min helped to reduce the leakage current substantially while in use, as compared to the detectors fabricated following conventional methods.<sup>43</sup> Thus, surface cleaning and modification with **CP** for a few minutes in various application areas are innovative, quick, cost-effective, and eco-friendly.

In summary, some of its salient features include (1) A Single-step, solvent-free approach; (2) Deposition of the organic, inorganic, nanocomposite, nanoparticle coating possible (e.g. polymer, silica-like coating, anti-scratch films, ceramics *etc.*); (3) Typically polymeric package materials may be suitably modified by increasing the surface energy to facilitate glueing and to improve printability, avoiding the need of using a primer, promoting coloring (dyeing); (4) Improvement of mechanical properties, added *hydrophilicity* or *hydrophobicity*, reduced shrinkage, *etc.*; and (5) Surface or Interface cleaning. *In this context, it may be a logical and appealing proposition to try and test if diamonds' surfaces get colored from the introduction of controlled surface defects by CP and thus mimic NR, which is used for creating artificial (bulk volume) color in commercial diamonds.*

**E. CP in Food Technology and Related Applications:** For many decades in our country (as also adopted in some other countries), NR technology has been widely and effectively employed in the *Radiation Processing of Food Items*.<sup>44</sup> The pathogen and parasite free treated food gains in its shelf life and gets enabled for mere chilling in place of deep freezing, thus saving on energy. For example, exporting local fruits such as mangoes (to the USA) has become possible. However, the related safety conditions for NR use discussed previously need to be strictly adhered to while employing this utility and accepting its high cost and associated factors for using such an industrial facility.

In the context of **Food Items' Processing and Preservation**, it may be noted that the harmful pathogens, parasites, and pesticides reside mainly on or near the item surface. Hence, CP applications, with their rich contents of ROS/RNS discussed previously, may have a direct bearing on the objective. Examples of the use of CP treatments include the ongoing research in various countries, which provide convincing reports of its local success. A study in the Glasgow University in Scotland in 2013 revealed that various items inside a sealed package or non-metallic container without any additives could be exposed with CP wherein the Ozone produced within the box (type 1 in Figure 4) sterilizes the contents' exposed surface area to help extend product shelf-life, efficiently destroying or inhibiting the bacteria or virus growth that occur otherwise. A few examples of such treatment include reducing pathogens in poultry, healthy bread and muffins.<sup>45</sup> Similarly, another study revealed that with a few min of CP treatment, strawberries could be freed of microorganisms.<sup>46</sup> Another study from Purdue University revealed CP treatment helps extend the shelf life of tomatoes by 4 weeks.<sup>47</sup> In 2016, the New South Wales Department of Primary Industries and the Australian Government initiated research studies with CP deployment to benefit the growers, processors, packers and consumers of fruits (apple, blueberry, citrus, strawberry), nuts (macadamia, almonds), vegetables (spinach, lettuce, fresh-cuts), aiming to mitigate the food safety risks.<sup>48</sup> In 2018, the Vietnam Institute of Technology has initiated research on environment-friendly CP technology for decontamination of agricultural products and fresh food and vegetables.<sup>49</sup> Thus, incorporation of CP treatments in Food Industry and Technology, as also projected by a currently available commercial system,<sup>50a</sup> describes achievable benefits world over such as (a) Increased germination and yields; (b) reduced bacterial levels at harvest, especially with the use of CP generated nitrate fertilizer; (c) Degradation of various types of pesticides before storage and consumption; (d) Pest and mycotoxin removal; (e) Pasteurization and sterilization; and (f) most significantly, anticipated reduction of food wastage.<sup>50b</sup>

A recent monograph written and edited by experts in the field aims at academic researchers, food scientists, and

government officials working on the disinfection of food products. It unequivocally states that **Cold Plasma**, one of the newest and novel technologies for food preservation, is promising and affordable, waterless, waste-free, and leaves no chemical residue on the products. It works as a disinfectant of food products and packaging materials while maximizing microbial and spore inactivation.<sup>51</sup> Finally, this section would remain incomplete without the following anecdote from space. In 2001, experiments were funded by **The European Space Agency** to eliminate "*The foul exhaust air from cooking French Fries on the International Space Station*". CP did not fail to provide the most appropriate response to this issue. *A suitable device was installed that works by using electrons and Free radicals within the CP disc to neutralize odors. The CP breaks up the offending molecules into harmless components that do not smell and do not need to be extracted afterwards. The cleaning was found to be almost a thousand times faster than the traditional existing chemical method.*<sup>52</sup>

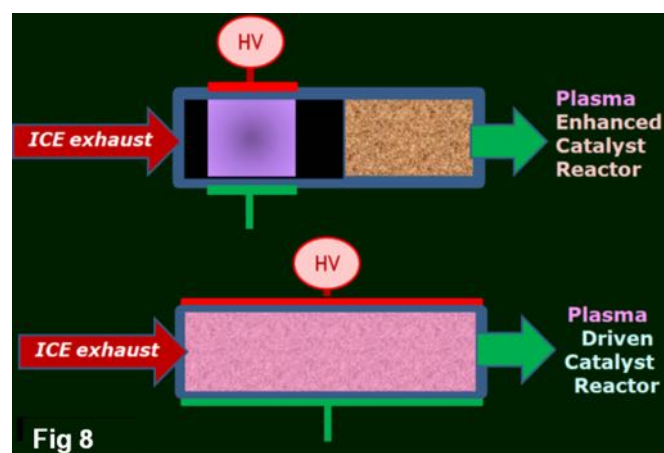
**F. CP in Travel and Transportation:** In the modern-day fossil-fuel driven transportation, a random comparison of two passenger car models (of two-door range) made by one manufacturer over a gap of almost 113 years, one finds the petrol mileage has barely improved, from reported **mpg** of 24 or 10 km L<sup>-1</sup> in 1908 to just 30 or 12 km L<sup>-1</sup> today. Of course, the overall improved getup and looks have not failed to impress. Similar mileages are also ascribed to other makes and models. To realize the prevailing scientific premise or lack of it leading to this puzzle, one needs to peek into various chemical steps that drive the internal combustion engines (ICE) and take a quick look into various transformations.

The fossil fuels employed in ICEs include either Petrol/Gasoline (C<sub>5</sub>-C<sub>8</sub> HCs) or Diesel (C<sub>10</sub>-C<sub>21</sub>) with ~ 86% C & 14% H contents. Taking the typical example of petrol, for its complete combustion, *ca.*  $2C_8H_{18} + 25O_2 \rightarrow 18H_2O + 16CO_2$ , each C<sub>8</sub>H<sub>18</sub> molecule needs 12.5 O<sub>2</sub> molecules to produce 9 H<sub>2</sub>O and 8 CO<sub>2</sub> molecules. The accurate picture regarding the combustion products is, however, quite different. The variety of products exiting the exhaust tailpipe, in addition to N<sub>2</sub>, CO<sub>2</sub> and H<sub>2</sub>O, include CO, NO<sub>x</sub>, NH<sub>3</sub>, SO<sub>x</sub>, RH, RR'H, C<sub>6</sub>H<sub>6</sub>, ROH, HCHO, RCHO, RCOOH, RCOR', RNH<sub>2</sub>, ArNH<sub>x</sub>, PAHs, VOCs, H<sub>x</sub>NO<sub>y</sub>, PM<sub>2.5</sub> & 10, Metals, etc.,<sup>53</sup> which suggests the oxidation reaction above remains nearly a fantasy, and occurrence of a large number of other unneeded chemical reactions, seriously affecting the anticipated mileage. (*In a related concern, it is no secret that fossil fuel usage contributes towards global warming, and in the persistence of localized harsh environmental pollution world over.*<sup>54</sup>) A back of the envelop random analysis for a car (with 4 cylinders, capacity 1 L each, with 2 fillings for 1 crankshaft revolution, running at 2400 rpm, at 50 kmph speed and top mileage of 10 km L<sup>-1</sup>) reveals a duration of a few ms (~ 12.5) available for the fuel oxidation, which is not sufficient for the oxidation reaction to getting to completion. Moreover, since

HC fuel oxidation needs only oxygen (just 21% of the air), the major 78% N<sub>2</sub> plays no role. No wonder, finally, all are waking up to this grim realization and seeking better alternatives. However, it is believed that fossil fuel usage may continue for a long and indefinite period; thus, appropriate correction in the fuel usage steps have become mandatory, forcing intensive *D<sup>4</sup>* (*design, development, demonstration, deployment*) in various directions. CP plays an active role in these applications, and the following details highlight its functional roles.

In the year 2007, research in the **Los Alamos National Laboratory, USA**, found that CP assisted fuel combustion (CPAC) is indeed a fertile area for R&D. An atomized fuel (RH) stream before combustion when passed through a CP zone attached downstream to an existing fuel injector, randomly broke down the long chains of hydrocarbons into smaller fragments (*typically at the energy input of < 10 eV, each molecule produced neutral Free radicals, R<sup>•</sup>/H<sup>•</sup>, while at > 10 eV various ionic Free radicals, R<sup>•+</sup>/R<sup>•</sup>H<sup>•+</sup>were formed*). Consequently, the combustion was stabilized, and the fuel-burning efficiency was also enhanced to raise *mpg* and reduce pollution.<sup>55</sup> In related research in 2018, the **Eindhoven University of Technology, the Netherlands**, in their study dealing with *Cold Plasma Flame Diagnostic & Modeling* revealed that the unique nature of Cold Plasma makes fossil fuel combustion leaner and cleaner and simultaneously minimizes greenhouse gas emissions.<sup>56</sup> In their study, a new CP based burner geometry allowed thorough experimental and theoretical investigations of the fundamental mechanisms. It revealed further optimization was necessary for the complex physicochemical parameters of CP flames in varied scales in space and time. Interestingly, at the same time in **Japan**, an engine attachment, Plasma Direct™ unit was commercialized and is available for various models of motorcycles, passenger car and SUV engines running on petrol/gasoline.<sup>57</sup> Placed upstream, it created a variety of *Free radicals* in the air-fuel mixture inside the ICE, which modified the combustion to reach all corners of the ICE, producing improved ignition and higher *mpg* values, in addition to being environmentally friendly. In the last few years, **Transient Plasma Systems in the USA** have developed several CP based products, including a *Nanosecond Pulsed Power Ignition System*, which allow many desired fuel ignition benefits.<sup>58</sup> In summary, CP assisted Fuel Reforming and Ignition helps to break down the long chains of hydrocarbons in fuel into smaller fragments, thus reducing the fuel ignition/oxidation time, improving the engine performance by ~ 20%, in addition to the reduced presence of unburned and undesirable combustion products such as CO, NO and NO<sub>2</sub> while utilizing less power than conventional *Spark Plug*. Simultaneous progress in electronics allowed matched generation of nanosecond pulse DC voltage with compact and portable hardware. However, the story of CP's contribution to transportation is not over yet. Keeping in view the world-over demands for a cleaner environment, purification of ICE exhausts

with CP are also being researched, with appreciable success, and more are on the way. Typically, as shown in **Figure 8**, the exhaust gas mixture is treated in a catalytic reactor (CR) chamber placed downstream to the ICE for treatment. Such deployment of CP in either of the two arrangements is possible: (i) Plasma-enhanced CR, wherein CP generation is sustained in the exhaust gas mixture to chemically activate it before the CR, or (ii) Plasma Driven CR, wherein CP is generated within the CR volume. For almost two decades, various studies have revealed the benefits of CP use in various combinations configurations, and constant and suitable modifications are in progress.<sup>59</sup> Thus, the key benefits achieved so far with CP based ICE exhaust gas treatment include (i) Oxidation of leftover or unburnt fuel and consequent reduction of its atmospheric release; (ii) Efficient and improved NO<sub>x</sub> / NH<sub>3</sub> transformations, mainly to N<sub>2</sub>; (iii) Applicability to the full range of Diesel and Petrol engines; (iv) Efficiency not being affected by sulfur content in fuel; (v) Various gaseous species released are N<sub>2</sub> H<sub>2</sub>O, O<sub>2</sub>, CO<sub>2</sub> even at low-temperature operation (cold start), with Plasma system power consumption ~1 kW *vis-à-vis* typical engine power of 60 kW (< 2%). Thus, from fuel input to tailpipe exhaust, CP can play varied roles and contribute to improvements in vehicle performance and the surrounding environment.



**G. CP Elsewhere:** The presentation above unveils various features of *Free Radical* Chemistry that occur within **Cold Plasma** and the related ease of system design, handling, use, and applications in a myriad of human activities, directly and indirectly. However, the story of its functional role would remain incomplete if the other related applications were not highlighted here. These include (a) Sterilization of medical devices on-site, which otherwise need to be autoclaved or N<sub>2</sub>O treated. The medical devices are sealed into a bag or tray and placed inside the CP device.<sup>60</sup> The device generates O<sub>3</sub> inside the bag to sterilize the device. The packet may be stored before use later. Excess ozone quickly decays back to oxygen, preventing any exposure of the user. (b) Printing μm sized

structures using nano inks by combining high-resolution plasma printing with rotogravure printing.<sup>61</sup> (c) Mercury-free Fluorescent Light, produced efficiently employing the reaction  $\text{Xe} - \text{CP} \rightarrow \text{Xe}^{*/++} - \text{Xe} \rightarrow \text{Xe}_2^* + \text{Xe}_2^{*+}$ , wherein xenon (in neon) discharge emission at  $\lambda = 147$  &  $172\text{nm}$  gets converted to white light with phosphor coating.<sup>62</sup> (d) Similarly, Plasma TV utilizes Xe (in Neon) CP, and the three primary colors Red/Blue/Green in pixels are generated with different phosphors.<sup>63</sup> (e) Plasma Clean Room Air Conditioner, working on the principle of air sterilization and cleaning discussed in the previous section C.<sup>64</sup> (f) A new Barrier Ionic Discharge (BID) detector in Gas Chromatograph employs a CP in He gas for sensitive and hassle-free universal detection and analysis of trace organic and inorganic compounds.<sup>65</sup> (g) For effective and quick removal of unpleasant odors from textiles, a portable and small hand-held device offers style and comfort at home and during travel, as it instantly refreshes the cloth without washing or deodorants.<sup>66</sup> In essence, the CP science and technology has conclusively proved that it is quite powerful, versatile, and can participate as a **standalone multidisciplinary approach** with many roles and applications, way beyond its projected role of mimicking the equally adaptable NR. *Finally, this presentation would remain unfinished without mentioning the phenomena giving rise to the **multihued, transcendental, ethereal Aurora (Figure 9)** observed in our sky near the poles (Aurora Borealis near the North Pole, and Aurora Australis near the South Pole). The vivid colors therein are generated by de-excitation of the excited oxygen atom (emitting green, some brownish red, yellow photons) and the nitrogen atom (blue, red, violet photons) as a CP in Giga-scale forms due to ionization of earth's upper atmosphere by charged particles released periodically within the solar wind.*<sup>67</sup>



**Cold Plasma** science and **Cold Plasma**-enabled technologies have thus revolutionized the modern society lifestyle and enabled our understanding beyond the fundamental processes that dominate the solar system and the interstellar

medium. It is hoped that with proper stewardship, its awareness for universal applicability would continue to grow and bring in new and innovative R&D the world over and simultaneously translates into pan India societal benefits to prove its true worth.

#### Author's note:

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† Few randomly selected publications from vast research contributions of Prof. S. R. Mohanty include: (a) S. R. Mohanty, A Relationship between Heat Conductivity and Viscosity of Liquids. *Nature* **1951**, 168, 42. <https://doi.org/10.1038/168042a0>; (b) S. R. Mohanty, S. Upadhyay, Influence of Crystal Defects on the Annealing of Chemical Radiation Damage. *Nature* **1963**, 199, 169. <https://doi.org/10.1038/199169b0>; (c) S. R. Mohanty, S. Upadhyay, Annealing of Chemical Radiation Damage by Compression. *Nature* **1964**, 201, 921. <https://doi.org/10.1038/201921a0>; (d) S. R. Mohanty, S. Nair, Photo-annealing of Chemical Radiation Damage. *Nature* **1967**, 215, 1476-1477. <https://doi.org/10.1038/2151476b0>.

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