

PREFACE

Special issue on environmental applications of thermal and non-thermal plasmas

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Preface

Special issue on environmental applications of thermal and non-thermal plasmas

Guest Editors

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It is our great pleasure to present this special issue of *Journal of Physics D: Applied Physics* dedicated to the general subject of environmental applications of thermal and non-thermal plasma. Plasma processing capabilities have a depth and breadth of versatility that can perhaps be best appreciated when considered from the perspective of environmental applications. Since the first development of ozonization and electrostatic precipitation more than 100 years ago, the environmental applications of plasma have broadened considerably to include removal of air pollutants such as nitrogen oxides (NO_x), sulfur oxides (SO_x), methane (CH₄), and volatile organic compounds in the exhaust gas stream of stationary or mobile sources, carbon dioxide (CO₂) conversion and water treatment as well as other areas. Hundreds of papers on the topic of environmental applications of plasmas have been published and this special issue presents 17 of the latest advancements in selected areas of environmental applications of thermal and non-thermal plasma.

One of the oldest environmental applications of electrical discharges is in the field of electrostatic precipitation (ESP). While traditionally applied in industries involving the use of utility boilers and cement kilns, ESP has recently been investigated for many new applications including cleaning of diesel exhaust and of indoor air in houses and hospitals. In their contribution to this special issue, Li *et al* investigated the effects of voltage rise on the performance of lab-scale and industrial electrostatic precipitators, in particular spark-over voltage, voltage recovery time following a spark-over, and gas flow patterns [1]. The work shows that the inlet fields of an ESP are mostly operated near the spark-over voltage. A comparison of different power sources reveals that a three-phase transformer/rectifier performs much better in terms of high voltage power consumption and recovery time than single-phase and high frequency power sources. Flow imaging studies further demonstrate that corona discharge-induced vortexes can be utilized for improving the fine particle collection. To avoid the production of ozone and improve the energy efficiency of electrostatic precipitation, Takashima *et al* investigated the feasibility of induction charging as an alternative method of charging particles in a two-stage ESP [2]. Results show that airborne particles are able to be charged in this way without corona discharge generation. However, the charging characteristics appear to be significantly affected by the electric field strength and air flow in the induction chamber, so further work is needed to improve the energy efficiency of induction charging under high air flow conditions.

To treat contaminated circulating indoor air, Timmermann *et al* developed a novel device that is based on a combination of a surface dielectric barrier discharge (DBD) and ionic wind [3]. The system is highly effective in inactivating *Escherichia coli*, the airborne test bacteria, achieving 20% removal by the plasma treatment alone and up to 90% removal by the combined system while producing only 360 ppb of ozone. The surface DBD appears to be strongly influenced by the gas humidity which has been shown to affect power consumption, discharge expansion on the surface, and ozone production. Overall, the preliminary results are encouraging and warrant further investigation into the operation and scaleup of this and similar devices.

Plasma-based water treatment (PWT) has been studied since the 1980s. The process has been applied to the treatment of numerous compounds including pharmaceuticals, organics dyes, phenols, and other emerging and legacy contaminants [4]. Five contributions to this special issue are devoted to PWT with topics ranging from plasma reactor design and treatment effectiveness to challenges and solutions for process scaleup.

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In this special issue, Magureanu *et al* provide an extensive literature review on the removal of pesticides in water by plasma treatment [5]. The authors indicate that the research shows that over a variety of reactor geometries, the energy efficiency of the treatment depends on the mass transfer of reactive species from the plasma to the liquid. Despite achieving excellent removal efficiencies as a standalone process, their review suggests that for the most effective elimination of water pollutants, plasma treatment may have to be combined with additional advanced oxidation processes such as ozone and Fenton's reaction.

Today, one of the most promising plasma reactor designs in terms of scaleup and energy efficiency involves generating plasma in a gas phase contacting a liquid. In another article, the performance of one such reactor for the destruction of herbicide metolachlor under three different discharge regimes (DBD, DC- and DC+) was investigated by Nani *et al* [6]. Measurements of the production rates of three reactive oxidative species (ROS); OH radicals, ozone and hydrogen peroxide suggest that different modes of power delivery control the production rates of ROS and thereby the treatment efficiency. This finding is of enormous importance for the further development and scaleup of electrical discharge plasma reactors, in particular optimization of the plasma-generating electrical network. The results of this work are also expected to inspire future similar studies since investigations on the influence of the mode of energy deposition on plasma chemistry are scarce.

In the contribution by Krupež *et al*, an aqueous solution containing nicotine was treated in a water falling film DBD reactor [7]. In drinking water treatment plants, nicotine undergoes chlorination during the disinfection process resulting in the formation of toxic byproducts. Thus, the primary goal of the study was to find the conditions under which nicotine can be completely degraded by non-thermal plasma treatment. Results showed that in terms of biodegradability, toxicity, and extent of degradation and mineralization, the most successful treatment was the direct plasma treatment in the presence of ferrous ions. The direct treatment resulted in 90% degradation over one hour of treatment. 13 nicotine degradation products were identified, all of which were generated following the oxidative transformation of the pyrrolidine ring.

Liu *et al* report a novel in-liquid arc plasma jet designed to treat contaminated water. The system uses a conventional arc torch to inject a plasma jet into the liquid [8]. One important feature of the system, as demonstrated by high-speed imaging, is the enhanced interaction of the plasma zone with the surrounding water, resulting in increased mass transfer of reactive species across the plasma-liquid interface. The thermal and electron impact reactions in the plasma were mainly responsible for the production of OH, O, H and nitrogen-based reactive species, although significant electrode erosion was also observed. The degradation of phenol, the model contaminant, showed that as specific input energy (SIE) is increased, the phenol degradation first increases rapidly to ~90% at $\text{SIE} = 1000 \text{ kJ l}^{-1}$, then slows down to show asymptotic behavior as it approaches 100% at which the value of SIE becomes greater than 3700 kJ l^{-1} . Oxalic acid was identified as the final degradation product.

Despite the significant progress that has been made in understanding the role of the reactor design in the effectiveness of plasma water treatment, the process is still limited to laboratory use. Foster *et al* offer an in-depth insight of the key challenges preventing the widespread implementation of plasma reactors for industrial water treatment [9]. In terms of plasma reactor design, the most effective plasma reactors are those in which the contact between the plasma and the treated solution is maximized and mass transfer limitations minimized. The authors present and discuss two scaleup design approaches and offer concrete steps for lab-to-field plasma reactor transition. To facilitate the comparison of the efficiency of plasma reactors for water treatment, the use of a standardized performance parameter, the electric energy per order, EEO, has been recommended.

The interest in the applications of nanosecond discharges in the areas of biomedicine, water sterilization, and water decontamination has increased tremendously in the last couple of years. In this special issue, Wang *et al* investigate the physics and chemistry of argon and helium pulsed nanosecond discharges propagating along the surface of a water film [10]. The work highlights the connection between the basic operational parameters such as discharge power and the resulting chemical and electronic processes in the plasma and the surrounding liquid. While many studies investigate the plasma region separately from the bulk liquid region, this work is a step forward in the development of a more comprehensive picture of the chemical processes occurring at the plasma-liquid interface.

In their contribution to this special issue, Takehana *et al* used a (water) thin film non-thermal plasma reactor to remove nitrogen oxides, sulfur oxides and nanoparticles from air [11]. Under certain conditions, nanoparticle collection efficiencies were $>99\%$ while the removal efficiencies for both NO_x and SO_x exceeded 97% . The authors also investigated the role of flowing water on nitrogen chemistry and found that the presence of water (i.e. OH radicals) is required for NO generation.

Another niche application of electrical discharge plasma lies in the removal of tar from syngas produced by gasification of fuel or biomass. The gasification process, due to the very high temperatures involved, generates significant levels of tars and particulates with syngas which are problematic in integrated biomass gasification systems. In their special issue paper, Cimerman *et al* investigate tar (naphthalene) removal by atmospheric pressure DBD in combination with various packing material, and present the conditions for achieving the highest removal and energy efficiencies (i.e. oxygen plasma in combination with a TiO_2 catalyst) [12]. Their in-depth analysis of the gaseous and solid products formed during the process reveals a rich and complex plasma chemistry with reactive oxygen species having a dominant role in the oxidation of naphthalene. The comparison of their results with other published studies reveals that the non-thermal plasma process should be combined with a catalyst to achieve the highest process removal and energy efficiencies.

Many of the contributions to this special issue used some form of a catalyst to improve the process efficiency. As demonstrated in past studies, the potential of a catalyst to control the plasma chemistry is rather large but it requires a profound understanding of the nature of the interactions between the plasma and the catalyst [13, 14]. Kim *et al* used intensified charge coupled device (ICCD) imaging to investigate the effect of negative polarity discharge on plasma generation and streamer propagation over the surfaces of titanium dioxide (TiO_2) and alumina ($\gamma\text{-Al}_2\text{O}_3$) in a packed-bed reactor [15]. Unlike for the positive polarity discharge, primary streamers were not found for all experimental cases tested in the study. Instead, glow-like discharge appeared at the cathode in the early stages. After its disappearance, counterpropagating cathode-directed streamers were initiated from the ground electrode (anode). Their formation was explained by a numerical model and their appearance captured in detailed time-resolved ICCD images. The morphology and dynamics of cathode-directed streamers were found to be strongly influenced by the presence of oxygen.

The interaction between a DBD and Ni-supported Al_2O_3 for dry methane reforming was investigated by Kameshima *et al* [16]. To elucidate unique reaction pathways induced by the discharge, coke formation behavior and morphology were investigated. Analyses revealed that the interaction between the plasma and the catalyst occurs exclusively at the catalyst's surface. Neither generation of DBD nor the diffusion of plasma-generated reactive species in the internal micropores is possible. Coke formation behavior has been explained by the catalyst effectiveness factor, that is, considering surface reactions rates and pore diffusion rates. Plasmochemical reactions resulting in coke formations are also discussed.

Nanosecond repetitively pulsed discharges that operate at atmospheric pressure are currently of great interest as a fuel-reforming technology. In their contribution to this special issue, Maqueo *et al* characterized such a device for partial oxidation of methane using a pin-to-mesh electrode configuration [17]. Experiments revealed the existence of two stable discharge regimes: low power diffuse and more energetic filamentary regime. The two regimes, which have been shown to depend on the applied voltage, differ significantly in electron number density, rotational and vibrational temperatures and chemical yields. To produce syngas, Brune *et al* investigated dry reforming of methane in a packed bed DBD at atmospheric pressure [18]. In the presence of a catalyst, conversions of both, CO_2 and CH_4 increased. Furthermore, up to 90% conversion of CH_4 from a CO_2/CH_4 mixture has been achieved using a copper-containing catalyst. Surface characterization measurements revealed that copper is predominantly present in a metallic state on the surface of gamma Al_2O_3 which influences the electrical characteristics of the discharge and allows for better adsorption and desorption of atomic oxygen and carbon monoxide on copper, compared to nickel and cobalt catalysts.

The special issue also features several excellent contributions on the topic of plasma-assisted CO_2 conversion. Considering the recent renewed scientific interest in the subject, they are indeed timely. Britun *et al* discovered significant improvements in the energy efficiency of CO_2 conversion by optimizing the plasma pulse repetition rate and gas velocity

during a surfaguide microwave discharge [19]. In addition to improving the actual conversion efficiency, the authors offer fundamental insights into the relevant energy transfer mechanisms and explain how those may be affected by power modulation. Lu *et al* investigated the decomposition of CO₂ in a DBD reactor with and without a catalyst [20]. One important finding of this work concerns the role of the gas temperature in the plasma reactor, which has been shown to influence the decomposition of CO₂ significantly, yielding energy efficiencies of up to six times higher for the temperature-controlled (cooled) plasma reactor compared to the non-cooled case.

We would like to thank all the authors for their excellent contributions and hope they inspire future investigations that may not only solve existing challenges but also create new applications of thermal and non-thermal plasmas to environmental concerns.

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