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First results on transient plasma-based remediation of nanoscale particulate matter in restaurant smoke emissions

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ABSTRACT

Recent studies have shown that nanoscale particulate matter produced in commercial charbroiling processes represents a serious health hazard and has been linked to various forms of cancer and cardiopulmonary disease. In this study, we propose a highly effective method for treating restaurant smoke emissions using a transient pulsed plasma reactor produced by nanosecond high voltage pulses. We measure the size and relative mass distributions of particulate matter (PM) produced in commercial charbroiling processes (e.g., cooking of hamburger meat) both with and without the plasma treatment. Here, the plasma discharge is produced in a 3" diameter cylindrical reactor with a 5–10 ns high voltage (17 kV) pulse generator. The distribution of untreated nanoparticle sizes is peaked around 125–150 nm in diameter, as measured using a scanning mobility particle sizer (SMPS) spectrometer. With plasma treatment, we observe up to a 55-fold reduction in relative particle mass and a significant reduction in the nanoparticle size distribution using this method. The effectiveness of the nanoscale PM remediation increases with both the pulse repetition rate and pulse voltage, demonstrating the scalability of this approach for treating particulate matter at higher flow rates and larger diameter reactors.

1. Introduction

During the past couple of decades, the adverse health effects of particulate emissions have been firmly established by many toxicological studies (Lighty et al., 2000; Burtscher, 2005; Brown et al., 2001; Oberdörster et al., 2004). In epidemiological reports, these ultrafine particulates have been linked to premature cardiovascular and respiratory deaths in metropolitan areas, as well as lung cancer (Samet et al., 2000; Pope et al., 2002; Chow et al., 2006; Oberdörster et al., 2005). A 1993 study published by Dockery et al. has been cited more than 4600 times as of the time of this writing, demonstrating the broad impact of this problem (Dockery et al., 1993).

Since 1997, the South Coast Air Quality Management District (SCAQMD) in Southern California has regulated smoke emissions from chain-driven (i.e., conveyor-belt) charbroilers under RULE 1138 (RULE 1138, 1997). These emissions consist of oil aerosol particles centered around 100–200 nm in diameter that are generated from the charbroiling of fat contained within the meat being cooked. In these chain-driven charbroilers, high temperature catalysts are placed just a few inches above the cooking surface and provide effective mitigation of the oil aerosol pollutants. These chain-driven charbroilers are typically found only in large fast-food restaurants. However, a vast majority of restaurant smoke emissions (~85%) originate from open-underfire charbroilers. In New York City, these open-underfire charbroilers emit an estimated 1400 tons of PM annually. The New York Department of Health estimates that more than 12% of the $PM_{2.5}$-attributable premature deaths can be attributed to these charbroiler emissions (New York City Administrat, 2016). If all restaurant charbroilers in the New York metropolitan area were equipped with pollution control technologies, a substantial number of these premature deaths could be prevented through reduced $PM_{2.5}$ concentrations.

It should be noted that the high temperature catalysts that are used for chain-driven charbroilers are not suitable for treating open-underfire charbroilers. Here, the exhaust hood is approximately 1 m away for the hot cooking surface. As such, the exhaust cools down substantially, by the time it reaches the hood, and would thus require additional heating of the catalyst in order to function properly. Dr. Karavalakis and coworkers at the University of California at Riverside has recently performed a comparative study of three pollution control technologies for removing PM from commercial meat cooking operations using the

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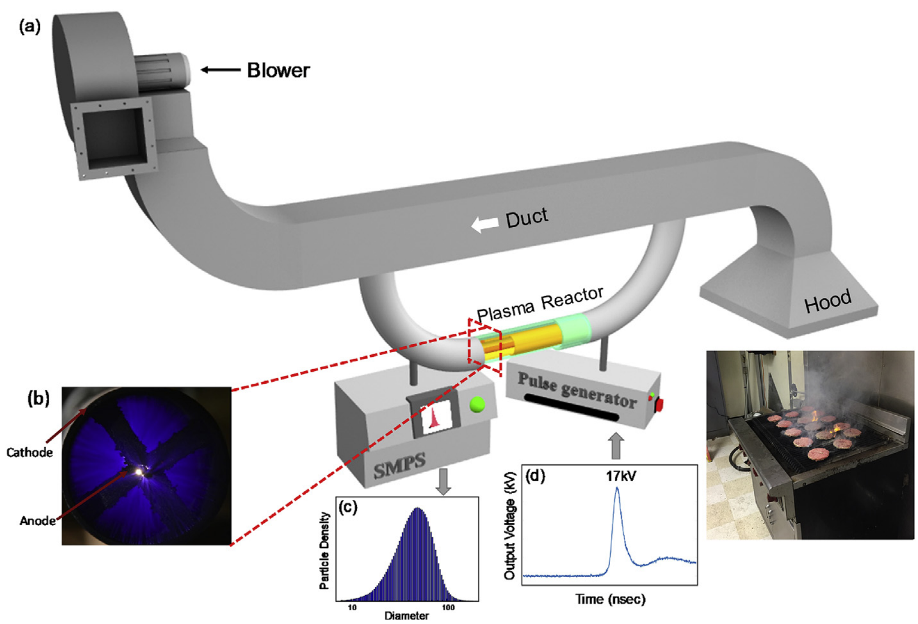

Fig. 1. (a) Schematic diagram of the experimental setup used to test the transient pulsed plasma reactor. In this configuration, the plasma reactor is installed in parallel to a kitchen ventilation system including a charbroiler, hood, duct, and blower. Here, only a fraction of the full flow is passed through the reactor. (b) Photograph of the transient plasma (high electron energy, low-temperature plasma), (c) typical particle size distribution, and (d) output characteristics of the nanosecond pulse generator.

South Coast Air Quality Management District (SCAQMD) Method 5.1 testing procedure (Gysel et al., 2018). These technologies include filtration, electrostatic precipitation (ESP) and steam injection. A similar study was carried out in Korea by Lee et al. (2011). For cooking applications that produce a large amount of grease particles (e.g., hamburger charbroiling), filter-based approaches become cost-prohibitive, as expensive filters must be replaced frequently. Also, with filter-based approaches, 2–3 filters are typically configured in series, resulting in a considerable pressure drop which, in turn, requires high power blowers to be utilized in order to achieve the necessary flow rates for kitchen ventilation compliance. The accumulation of grease in electrostatic precipitation systems also poses a potential fire hazard, and frequent cleaning of the collection plates is required.

2. Methodology

In the work presented here, we utilize a transient pulsed plasma to reduce nanoscale PM produced in a commercial charbroiling process. Here, the plasma-based flow reactor consists of a 3 ft-long, 3 inch-diameter stainless steel cylindrical anode with a 4-wire array of cathode center electrodes, as shown in Fig. 1. The overall footprint of the system is approximately 0.5’ × 3.5’. The plasma is produced using a TPS Model 20X pulse generator operating at a peak voltage of 17 kV, pulse repetition rates up to 2000 Hz, and continuous powers up to 80 W. A typical waveform from this pulse generator is plotted in Figure S1 of the Supplemental Information document. While radio frequency (RF)-based plasma reactors have been investigated for remediation of diesel exhaust for several decades (Guo, 2013; Chang, 2008; Bai et al, 2009; Dale et al., 1997; Di Natale et al., 2013; Kuwahara et al., 2012; Hackam and Akiyama, 2000), the nanosecond pulsed plasma used here consumes far less energy in the creation of the plasma. At a peak voltage 17 kV, our system delivers a transient power of 4.76 MW. The transient nature of the plasma necessitates that very little current is drawn in its creation. That is, once the streamer is created, the applied electric field collapses before a substantial amount of current (and hence electric power) can flow.

Remediation experiments were carried out at the Center for Environmental Research & Technology (CE-CERT) test kitchen in Riverside, CA. Here, the transient plasma reactor was installed in the CE-CERT facility, as illustrated in Fig. 1. Particle distributions were measured using a scanning mobility particle sizer (SMPS) spectrometer (TSI Model 3776) with a condensation particle counter (CPC) over the range from 14 to 685 nm. The scanning time of each dataset was 120 s while the aerosol flowrate of the SMPS was set at 0.3 LPM, and the sheath flowrate was 3 LPM. Hamburgers (75% lean, 25% fat) were cooked for 4.5 min per side continuously for 3 h during this study. 15 patties were cooked at a time on a grill that was 25” × 30” in area. A total of 375 patties were cooked during this study. Baseline particle distributions (i.e., histograms) were measured using the SMPS without a plasma exhibit highly stable distributions, as shown in Figure S2 of the Supplemental Information document.

3. Results and discussion

Fig. 2 shows the particle number densities measured with and without the plasma treatment for two different reactor flow conditions: 2.5 m/s and 0.25 m/s (790 and 79 LPM, respectively). For these datasets, the original untreated particle distributions peaked around 125–150 nm diameter. With plasma treatment, a significant drop in the particle number was observed along with the emergence of a narrow distribution centered around 30–40 nm. The integrated area of the dominant peak shows a factor of 1.7X reduction in PM number density (i.e., 4.62/2.71 = 1.7X) at high flow rates (2.5 m/s) and a 10-fold reduction in PM at low flow rates (0.25 m/s), as shown in Fig. 2b.

Since smaller diameter nanoparticles have substantially lower mass than larger diameter nanoparticles, it is more appropriate from a regulatory perspective to plot the particle mass instead of number density. Fig. 3 shows the relative mass density obtained by multiplying the particle number densities in Fig. 2 by the radius cubed. Here, we observe a 2.4- and 55-fold reduction in relative PM mass for flow rates of 2.5 and 0.25 m/s, respectively. In this representation, the narrow distribution of particles centered around 30–40 nm is negligible compared with the larger diameter particles, because of the diameter-cubed mass relation.

The nanoparticle distributions were also measured as a function of the pulse repetition rate. Fig. 4 shows the integrated particle number plotted as a function of pulse repetition rate, which decreases linearly with increasing repetition rate. Here, each pulse delivers approximately 40.2 mJ of energy. To first order we assume that the total power is proportional to the pulse repetition rate. These results demonstrate that this approach can be scaled up to treat higher flow rates at higher pulse repetition rates.

The particle distributions were also measured as a function of voltage dependence, as shown in Fig. 5. Here, again, a monotonic decrease

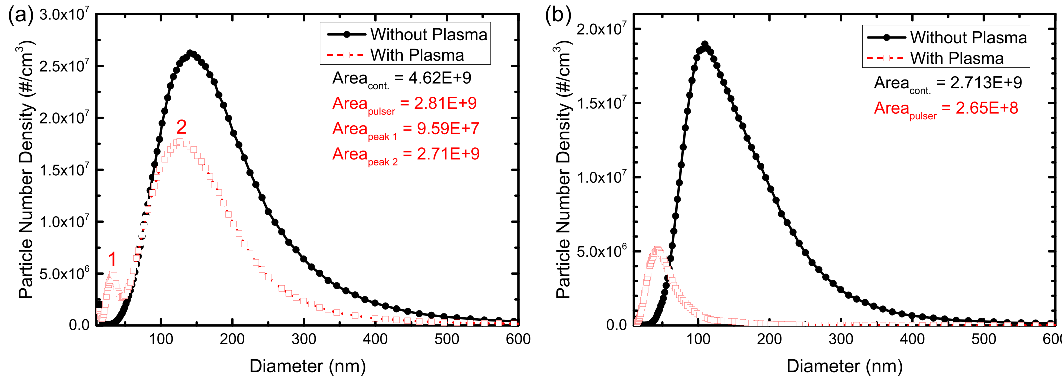

Fig. 2. Particle number densities measured with and without the plasma treatment for different flow conditions of (a) 2.5 m/s and (b) 0.25 m/s. The integrated peak areas are indicated in the figures. Here, the pulse generator was operating at a peak voltage of 17 kV, pulse repetition rate of 1200 Hz, and continuous power of 75 W.

is observed in the integrated area of the PM peak distribution (i.e., relative PM mass), with an overall reduction of 40x observed at a pulser peak voltage of 17,830 V. These input voltages correspond to pulse energies of approximately 10, 20, 40, and 50 mJ. These results further demonstrate the scalability of this approach for treating higher flow rates and larger diameter systems with higher pulse voltages.

Plasma-based treatment of diesel engine exhaust has been demonstrated by many groups for both PM and NOx remediation, including a large effort at the Ford Motor Company, nanosecond pulsed plasmas consistently outperform conventional RF-based plasmas. As mentioned above, this transient plasma draws very little current in creating the plasma since the applied electric field collapses once the plasma is formed and, thus, very little current (and hence electric power) can flow. Matsumoto et al. reported a comparison of the NO removal efficiency of nanosecond pulse discharge technologies with pulsed corona discharge and dielectric barrier discharge (DBD) reactors, which dissipate a substantial amount of energy as heat (Matsumoto et al., 2010). The nanosecond pulse discharge produces a “cold” plasma in which the electron energies are around 30 eV ($T = 10^5$ K), while the vibrational modes of the molecules remain close to room temperature. These highly energetic (or “hot”) electrons enable new chemical pathways through the formation of charge-free radicals and highly reactive species, including atomic oxygen and ozone, which are known to break down grease into CO, CO_2 and other smaller hydrocarbons (Goldstein and Kalk, 1981). These high reactive species drive chemical reactions that are fundamentally different from those of standard equilibrium chemistry. In addition, it is possible that the plasma induces the formation of smaller nanoparticles, which appears as a distinct peak in the spectra, corresponding to newly nucleated particles. Also, due to the limitations of our measurements (i.e., SMPS), we are unable to analyze the possible formation of PM with diameters smaller than 14 nm. The main

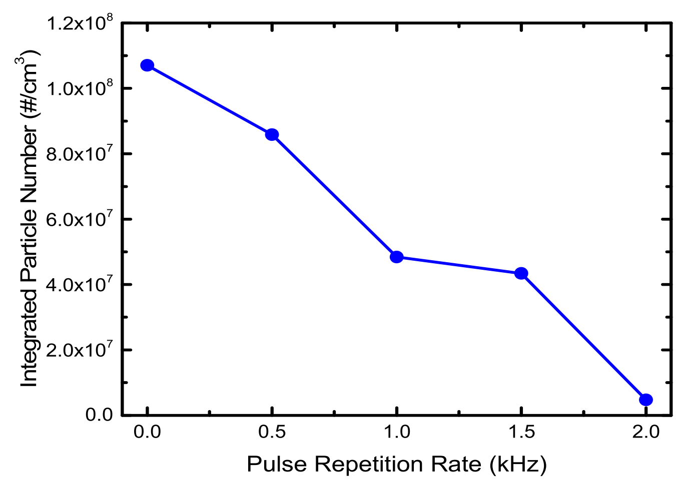

Fig. 4. Integrated particle numbers (i.e., total particle number) plotted as a function of pulse repetition rate taken under low flow conditions.

difference between plasmas created in diesel engine and restaurant exhaust is the temperature, which is less than 100 °C for restaurant smoke. While the same amount of energy is required to produce a plasma in both applications, the higher temperatures associated with diesel engine exhaust lead to arcing at lower thresholds, which ultimately limits the power/plasma density that can be achieved in these two applications.

4. Conclusion

In conclusion, these preliminary measurements show the

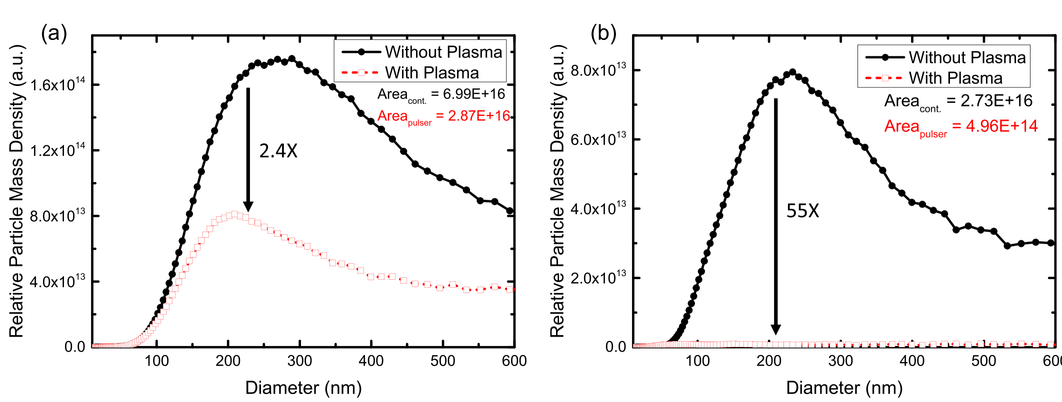

Fig. 3. Relative particle mass measured with and without the plasma treatment for different flow conditions of (a) 2.5 m/s and (b) 0.25 m/s. The integrated peak areas are indicated in the figures. Here, the pulse generator was operating at a peak voltage of 17 kV, pulse repetition rate of 1200 Hz, and continuous power of 75 W.

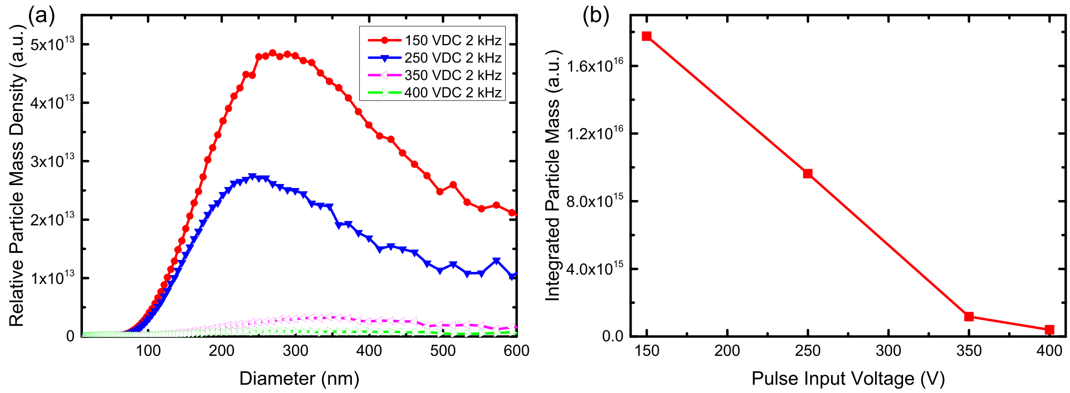

Fig. 5. (a) Particle number densities measured for various pulse generator input voltages (b) Integrated peak areas (i.e., relative particle mass) plotted as a function of pulse generator input voltage taken under low flow conditions.

effectiveness of transient pulsed plasmas to provide substantial remediation of PM produced by commercial charbroiling processes (e.g., cooking of hamburger meat). Using a SMPS spectrometer, we observe the distribution of untreated nanoparticle sizes to be centered around 125–150 nm diameter. A 55-fold reduction in relative particle mass is observed with plasma treatment, as well as a significant reduction in the nanoparticle size distribution. Here, the remediation of nanoscale PM increases with pulse repetition rate and pulse voltage, demonstrating that this general approach can be scaled up to treat higher flow rates and larger systems. This transient plasma-based approach provides a new method for breaking down oil-based PM that is fundamentally different from UV and/or ozone approaches, which are effective in treating odor but not PM (Rimatori et al., 1983; Hubbard et al., 2005; Li et al., 2009; Rifino et al., 1965; de Gouw and Lovejoy, 1998; Isaxon et al., 2013; Wang and Waring, 2014). Here, we believe that the formation of active free radicals in the plasma, such as atomic oxygen, break down the grease particles into CO, CO_2 and other smaller hydrocarbons similar to the mechanism by which plasmas break down polymer films (Goldstein and Kalk, 1981).

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envres.2019.108635.

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