Journal of Environmental Chemical Engineering 9 (2021) 106565

Contents lists available at ScienceDirect

Journal of Environmental Chemical Engineering

journal homepage: www.elsevier.com/locate/jece

Plasma-enhanced electrostatic precipitation of diesel exhaust using high voltage nanosecond pulse discharge

Sisi Yang [a], Indu Aravind [a], Boxin Zhang [b], Sizhe Weng [c], Bofan Zhao [c], Mark Thomas [d], Ryan Umstattd [d], Dan Singleton [d], Jason Sanders [d], Stephen B. Cronin [a,c,d,*]

[a] Department of Physics and Astronomy, University of Southern California, Los Angeles, CA 90089, USA
[b] Mork Family Department of Chemical Engineering and Materials Science, University of Southern California, Los Angeles, CA 90089, USA
[c] Ming Hsieh Department of Electrical Engineering, University of Southern California, Los Angeles, CA 90089, USA
[d] Transient Plasma Systems, Inc., Torrance, CA 90501, USA

A R T I C L E I N F O

Editor: Dr. G. Palmisano

Keywords:
Particulate matter
PM
Soot
PM2.5
PE-ESP
Pollution control device

A B S T R A C T

We have demonstrated nearly 80% remediation of diesel particulates from an 18 kW (i.e., 24.4 HP) diesel engine with just 1.6% of the engine power using a transient pulsed plasma in conjunction with an applied DC bias voltage. This result was achieved using a high voltage (~10 kV) DC bias together with nanosecond high voltage pulses (~20 kV) in what we refer to as a plasma-enhanced electrostatic precipitator (PE-ESP). Here, the remediation takes place in a two-step process in which the particles are first charged by the ions in the plasma and then they are swept out to the collecting electrode by the applied DC bias. The fast rise times (i.e., $dV/dt \approx 10^{12}$V/sec) associated with these nanosecond pulses produce a streamer discharge with ion densities that are more than one order of magnitude higher than conventional DC coronas. These extremely high ion densities produce the enhanced electrostatic precipitation observed here. In fact, little or no remediation was observed with the DC bias only or with the nanosecond pulses only, and it is the combination of these two applied fields (pulsed and DC) that produces the robust enhancement observed here. This general approach of using nanosecond pulse discharge to produce plasma-enhanced electrostatic precipitation opens up new degrees of freedom in the design of new and more compact ESPs.

1. Introduction

Over the past few decades, the negative health effects of diesel particulate matter (PM) have been well established by numerous toxicological studies [1–4]. In these studies, fine particulates (particles smaller than 2.5 μm) were linked to premature deaths associated with cardiovascular and respiratory disease in metropolitan areas, including lung cancer [5–8]. The broad impact of this problem is reflected by a 1993 study, which has been cited more than 5100 times [9]. The International Agency for Research on Cancer (IARC), which is part of the World Health Organization (WHO), declared diesel exhaust as carcinogenic to humans in 2012 [10]. More recently, Anenberg et al. investigated the air pollution-related health impacts of transportation-sector emissions from 2015 to 2020 [11]. They reported that port towns Seattle and San Francisco lead early deaths by twice the global average due to air pollution from their ports. Several technologies have been developed to remediate diesel particulates, such as wet scrubbers, diesel particulate filters (DPFs), and electrostatic precipitators (ESP) [12–14]. Much of this technology was developed in response to the increasingly strict air quality regulations, including those imposed by the California Air Resources Board (CARB), the U.S. Environmental Protection Agency (EPA), and local air quality air management districts (AQMD). The recent trend towards electric vehicles touting *zero emissions* has been successfully implemented in small cars and light duty vehicles. However, for high power applications including trucks, ships, and backup generators, diesel combustion will likely remain a dominant source of power, thus, requiring further development of improved pollution control devices. While diesel particulate filters have been successful in mitigating emissions from light duty and heavy duty vehicles, they produce an engine back pressure, which ultimately results in lower engine efficiencies. Furthermore, regenerating these DPFs through a high temperature baking procedure ultimately converts the carbonaceous PM to CO_2, contributing tons of carbon to the atmosphere every day. Wet scrubbers are not efficient enough to remove diesel particles from ship

* Corresponding author at: Ming Hsieh Department of Electrical Engineering, University of Southern California, Los Angeles, CA 90089, USA.
E-mail address: scronin@usc.edu (S.B. Cronin).

https://doi.org/10.1016/j.jece.2021.106565
Received 2 August 2021; Received in revised form 18 September 2021; Accepted 9 October 2021
Available online 12 October 2021

diesel exhaust and conventional ESPs are too bulky to be used for mobile sources, such as cars and ships.

Several research groups have explored the use of nonthermal plasmas (DBD, pulsed voltage) to treat diesel exhaust emissions including reductions in both particulate matter (PM) and NOx (i.e., NO and NO_2) [15–20]. These studies typically entail small laboratory-scale reactors treating a small fraction of the total engine exhaust flow (i.e., slipstream measurements) rather than treating the full engine exhaust. Remediation values of on the order of 1 g/kWh have been reported [16, 20]. For the 18 kW diesel engine used in our study, this would require 5 kW of electrical power, which is 28% of the engine power and, therefore, not feasible in a practical application. It should be noted that, in this nonthermal plasma approach, no DC bias was applied, and this approach does not lead to electrostatic precipitation.

In a separate set of studies, several research groups have explored the use of a conventional DC only electrostatic precipitators (i.e., without plasma) for treating diesel particulate matter [21,22]. Many of these reports entail small laboratory-scale reactors treating a partial fraction of the total engine exhaust [23–25]. For treatment of full engine exhaust, typical voltages range from 7 to 70 kV, and typical remediation efficacies range from 70% to 80% [13,26,27]. Saiyasitpanich et al. reported the use of a wet electrostatic precipitator using a DC voltage to treat diesel PM, however, at 0% engine load [13]. Using a two-stage ESP, Zukeran et al. were able to achieve 78% remediation after allowing the exhaust gas to cool to room temperature using a heat exchanger [28]. Crespo et al. reported remediation efficiencies above 85% after letting the exhaust gas cool to room temperature. While arcing voltage thresholds are much higher at room temperature, allowing the gas to cool to room temperature is not feasible for most practical applications. Our approach that uses high voltage nanosecond pulses together with a

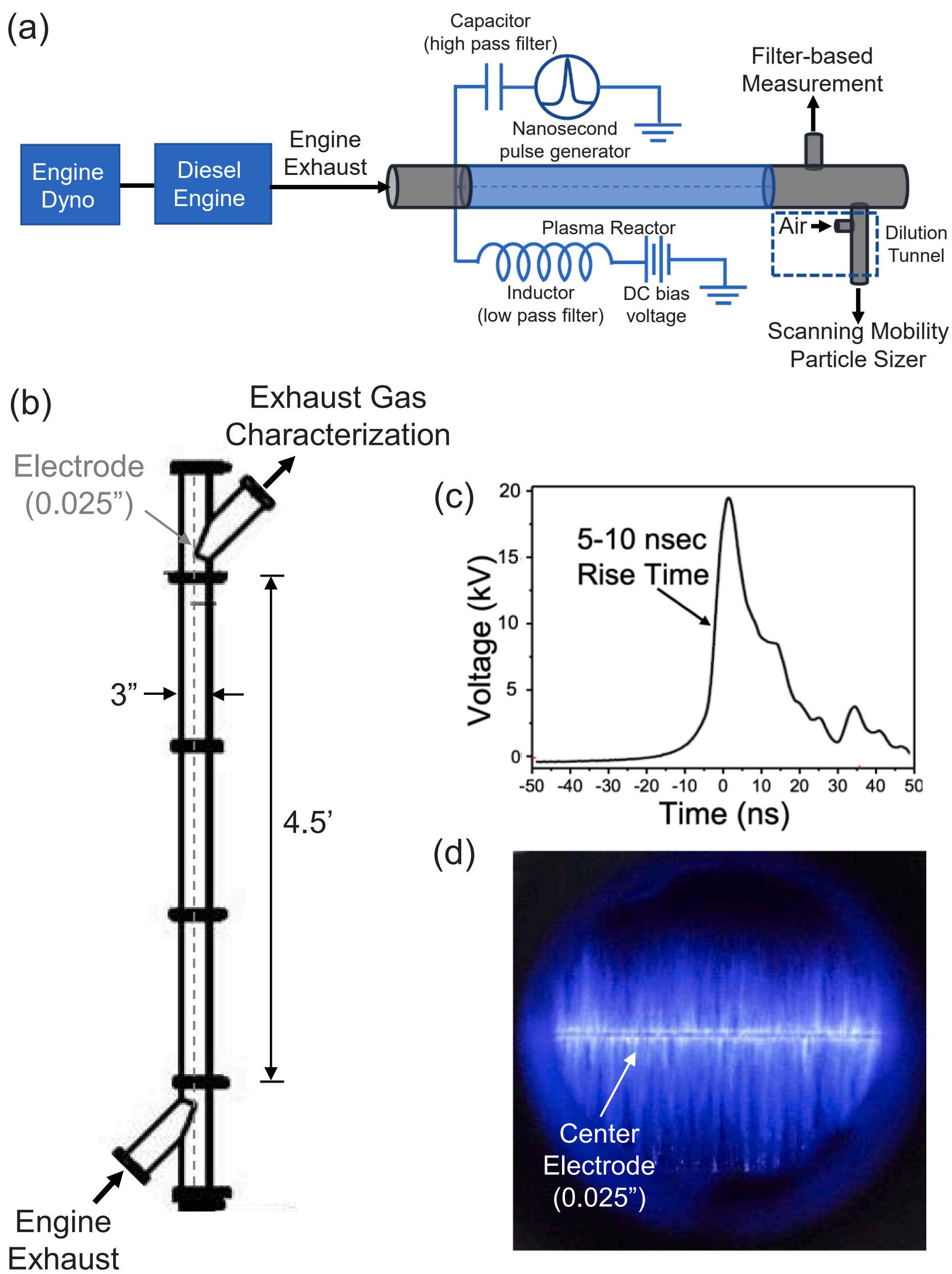

Fig. 1. (a) Schematic diagram of the measurement setup used for characterizing the plasma-enhanced electrostatic precipitator (PE-ESP) system. (b) Design diagram of the PE-ESP reactor. (c) Typical output characteristics of the nanosecond high voltage pulse generator. (d) Photograph of the transient plasma formed by high voltage nanosecond pulsed discharge.

DC voltage provides a way around this arcing limit, enabling higher peak fields and higher ion concentrations to be reached without arcing.

Several previous studies have explored the use of microsecond high voltage pulses to enhance electrostatic precipitators [26,29–31]. However, the use of ultra-short (nanosecond) pulsed plasmas to enhance ESPs has not been reported in the treatment of diesel exhaust [32,33]. One of main advantages of using nanosecond pulses over μsec pulses is power efficiency. Since the amount of energy per pulse is proportional to the pulse duration, a nanosecond pulse will be roughly 1000X more efficient than a μsec pulse with the same peak voltage. That is, once the streamer is formed in the nanosecond pulsed plasma, the applied electric field drops to zero before a significant amount of current can flow and power can be dissipated. This power vs. pulse duration relation is illustrated in Fig. S2 of the Supplement Document. In addition, the fast rise times associated with these nanosecond pulses (i.e., $dV/dt \sim 10^{12}$ V/s) give rise to a streamer discharge, as illustrated in Fig. 1d, which has a significantly higher (~20X) ion concentration than the corona discharge generated by DC voltages in conventional ESPs. This is particularly important in the high temperature exhaust gas environment, in which charged radical species have shorter lifetimes and arcing voltage thresholds are considerably lower. Rather than pulse duration, it is actually the pulse rise time (or more precisely dV/dt) that is proportional to the streamer density, including number of streamers per unit length of electrode wire and ion concentration produced within each streamer [34–37]. These streamers create extremely high peak ion concentrations that can enable more compact design of ESPs.

2. Materials and methods

Fig. 1 shows a schematic diagram of the experimental setup used for characterizing the plasma enhanced electrostatic precipitator. In this configuration, an 18 kW (i.e., 24.9 HP) diesel engine (Kubota, Inc., Model: V2203L-DI-EF01e, Family: EkBXL02.2RCB) is run under various load conditions from 0% (idle) to 100% load. The exhaust gases flow through the plasma reactor, which consists of a 3-inch diameter, 4.5-feet long metal cylinder with a 0.025-inch diameter wire in a coaxial geometry, illustrated in Fig. 1b. Also indicated in the Figure is the DC voltage supply used in conjunction with the nanosecond pulse generator with a high voltage capacitor high-pass filter and inductor low-pass filter. Fig. 1c shows a typical waveform of the high voltage nanosecond pulsed used in this study. Using this configuration, the exhaust gas temperature was 218 °C, the volumetric flow rate was 3260 lpm, linear flow rate was 11.9 m/s, residence time was 0.115 s, and the humidity was 20.6%. Our filter-based measurements were done following the ISO 8178 international standard test method for exhaust emission measurements. For the Kubota V2203L engine, we used a Type G2 test cycle, which is a 6-mode test including weighted averages over the following engine loads: 100%, 75%, 50%, 25%, 10%, and 0%. Further details of the exhaust gas conditions in these multi-mode tests are given in Table S1 of the Supplement Document.

Fig. 1d shows a photograph of the streamer discharge that is characteristic of these nanosecond pulses. The current-voltage characteristics of our system are plotted in Fig. S3 of the Supplement Document, and it shows that at $V_{DC} = 7$ kV there is minimal corona current, enabling the system to be operated well below the arcing threshold. Also, a substantially higher current is observed with nanosecond pulses used together with the DC voltage, reflecting the conductivity of the ions generated by this transient pulsed plasma.

3. Results and discussion

Fig. 2 shows the particle mass distributions of diesel exhaust measured using a scanning mobility particle sizer (SMPS) (TSI, Inc) both with and without the plasma-enhanced electrostatic precipitator treatment under 75% engine load with a dilution ratio of 8.1. In this Figure, the two black datasets show baseline data (i.e., with the plasma off and DC bias off) taken before and after all the other measurements. The red data set shows data taken with the DC bias only ($V_{DC} = 7$ kV) (i.e., without nanosecond high voltage pulses applied) showing no remediation. The blue dataset shows results obtained with both the DC bias and the nanosecond pulsed plasma on, exhibiting an 88.6% remediation. The apparent shift in particle size distribution to smaller diameters is likely due to the device being more efficient for larger particles, which are more easily charged and, therefore, more easily removed using electrostatic precipitation.

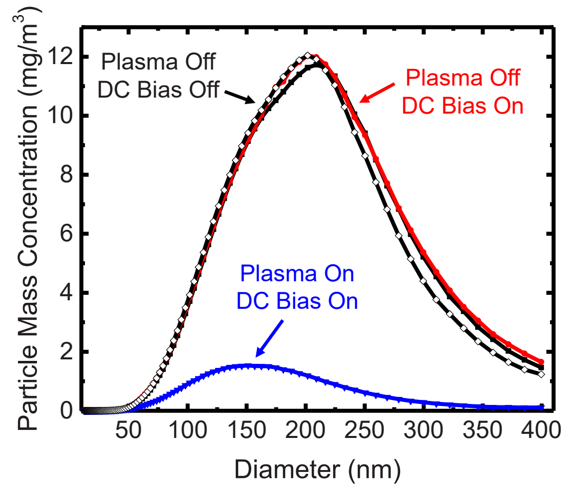

Fig. 2. Particle mass distributions measured in diesel exhaust with and without DC bias and nanosecond pulsed plasma.

Filter-based measurements remain the industry standard for regulatory emissions and enable us to collect data in grams of PM per brake-horsepower hour (g/BHP-hr). Here, we report the regulatory emissions standards carried out by a third-party testing facility Olson-Ecologic Engine Testing Laboratory (Fullerton, CA), which is an EPA and CARB-recognized engine testing facility. These tests were comprised of a Code of Federal Regulation (CFR) 1065–6 Mode G2 Test cycle, as defined by the U.S. EPA. In these measurements, the filters are weighed before and after flowing a controlled amount of engine exhaust through the filters using an AVL advanced dilution system. It should be noted that most previous studies of plasma-treated PM remediation were conducted by treating just a small portion of the exhaust flow (i.e., slip stream) [39–42]. However, our device treats the full flow and, thus, represents a major step forward in the practical treatment of diesel PM using electrostatic precipitation.

Fig. 3 shows the PM emissions caught using these filter-based methods in units of grams per brake horsepower hour (i.e., g/BHP-hr). Here, the untreated values are around 0.16 g/BHP-hr, and the plasma-enhanced ESP-treated values center around 0.035 g/BHP-hr. This data was reproduced in triplicate and resulted in remediation values of 79%, 80%, and 74%, and an average remediation of 78%. It should be noted that this remediation was achieved using only 273 W of electrical power (i.e., measured using a wall power meter), just 1.6% of the engine power using this plasma enhanced electrostatic precipitator technique. Based on these values, our PE-ESP removes approximately 3.1 g of particulate matter per hour corresponding to a removal efficiency of 11.4 g/kWh. This is one order of magnitude more efficient than the values reported by Fushimi and Okubo [16,20]. Further information of other exhaust gases (hydrocarbons, NO/NO_x, CO, CO_2, CH_4) under different engine loads are listed in Table S2 of the Supplement Document.

Fig. 4 shows the PM remediation plotted as a function of both peak pulse voltage and the applied DC bias voltage, as measured using an SMPS. Here, it is clear that both the DC bias and the nanosecond pulsed plasma are needed in order to provide appreciable remediation. For the data plotted in Fig. 4a, the peak voltage of the nanosecond pulsed

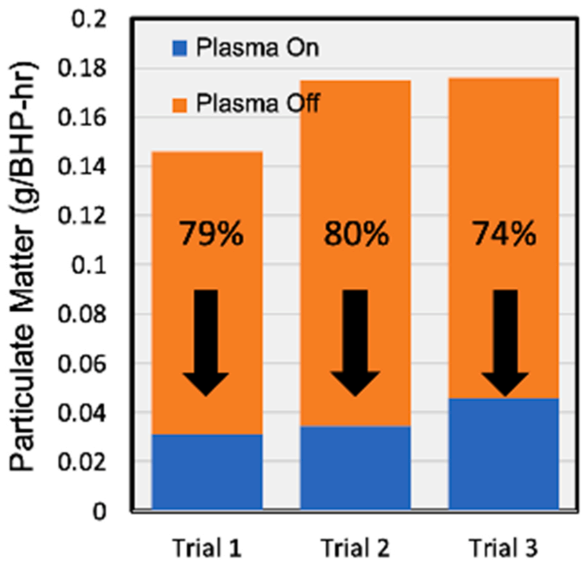

Fig. 3. PM emissions with and without plasma treatment of diesel engine exhaust performed on an 18.5 kW (24.9 HP) diesel engine showing an average remediation of 78% (PM in units of grams/brake horsepower hour (g/BHP-hr)).

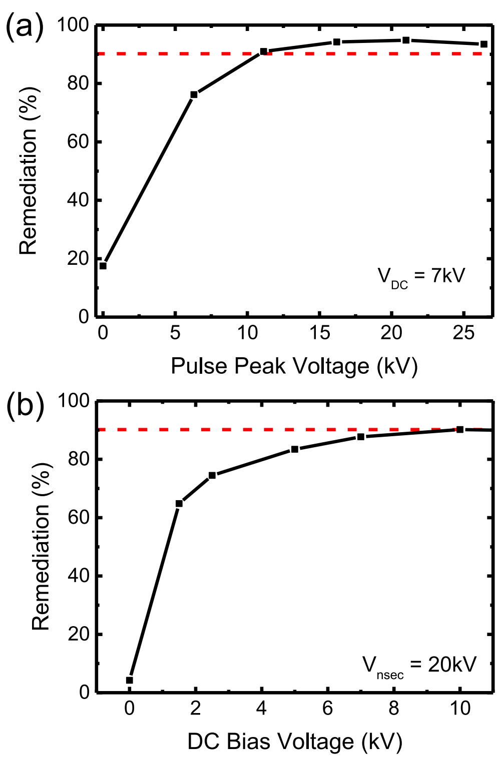

Fig. 4. Diesel PM remediation observed with an SMPS plotted as a function of (a) peak pulse voltage with a DC voltage of 7 kV and (b) DC voltage applied in conjunction with 20 kV nanosecond pulses at a repetition rate of 1 kHz.

discharge is swept from 0 to 25 kV, while a constant DC bias of 7 kV was applied. Here, the PM remediation increases sharply up to around 10 kV pulses, after which the remediation saturates. In Fig. 4b, the DC bias is swept from 0 to 10 kV, while a peak nanosecond voltage of 20 kV is held constant. Here, the PM remediation increases monotonically up until 10 kV, but begins to exhibit a saturation behavior beyond that point. Both plots show that remediation values of at least 90% can be achieved in these particulates. This is 10% higher than that which was observed by the filter-based method plotted in Fig. 3. Here, it is important to note that the SMPS measurements only captures particulate matter below 400 nm in diameter, whereas the filter-based measurements capture both nanometer and micron-sized particles. Therefore, in addition to electrostatic precipitation, it is likely that electrostatic agglomeration of these 200 nm diameter particles is occurring, thus, upconverting to micron-size clusters that are not being detected by the SMPS, but, do contribute to the filter-based measurements. We, therefore, believe that this 10% discrepancy in remediation is due to the different diameters with which these two measurement techniques are sensitive.

These voltage-dependent curves contain important information regarding the mechanism of plasma-based enhancement. The sudden increase in remediation between 0 and 5 kV pulse voltage (i.e., Fig. 4a) is due to the injection of ions at a DC bias that lies below the threshold for corona discharge/ionization. Below a pulse voltage threshold of 5 kV, there is insufficient ion density to achieve electrostatic precipitation. Similarly, below a DC voltage of 2 kV (Fig. 4b), there is insufficient electrostatic force to sweep out the charged nanoparticles within the limited residence time in the coaxial reactor. Fig. S3 shows the current-voltage characteristics of the PE-ESP system plotted as a function of DC voltage with and without nanosecond pulsed plasma. Here, we observe substantially higher current values with the nanosecond pulses indicative of the higher ion concentrations produced by the high voltage nanosecond pulses. The saturation behavior observed in Fig. 4a is likely due to re-entrainment associated with plasma-based ablation of soot material from the center electrode back into the gas phase. Since the plasma density is largest at that center electrode, where the electric field strength is maximum, we believe re-entrainment from the center electrode represents the key limiting factor for remediation in this current configuration of the PE-ESP. This is evidenced by the fact that the remediation saturates above peak pulse voltages of 10 kV, and even begins to decrease PM remediation at higher peak pulse voltages. In the collection of particulate matter using electrostatic precipitation, there is the inevitable question of what to do with the material that is collected? In our previous work, we have collected these residual soot particles from our reactor and used them as a conductive additive in cathode materials in Li-ion batteries [43]. This approach enables an abundant toxic pollutant to be converted into a valuable material for energy storage devices.

4. Conclusion

Here, we have demonstrated nearly 80% remediation of diesel particulates from an 18 kW (i.e., 24.4 HP) diesel engine with just 1.6% of the engine power using a transient pulsed plasma in conjunction with an applied DC bias voltage. This result was achieved using a high voltage (~10 kV) DC bias together with nanosecond high voltage pulses (~20 kV) in a plasma-enhanced electrostatic precipitator (PE-ESP) configuration. Little to no remediation was observed with the DC bias only or with the nanosecond pulses only. The nanosecond pulsed plasma used together with a DC bias provides a new way to make ESPs smaller. This approach will enable more compact ESPs to be developed, which could be potentially transformative in the mitigation of diesel particulates and deadly carcinogens. Thus far, ESP technologies have been limited in their use to large power plants (e.g., coal-fired power plants) that can accommodate large after treatment devices. Reducing the overall size of this technology, which has thus far been its main limiting factor, will open up new applications in mobile sources, such as ships and trucks.

CRediT authorship contribution statement

Sisi Yang: Methodology, Investigation, Data curation, Validation. **Indu Aravind**: Data curation. **Boxin Zhang**: Data curation. **Sizhe Weng**: Data curation. **Bofan Zhao**: Data curation. **Mark Thomas**: Investigation, Methodology. **Ryan Umstattd**: Investigation, Methodology. **Dan Singleton**: Investigation, Methodology. **Jason Sanders**: Investigation, Methodology. **Stephen B. Cronin**: Conceptualization, Investigation, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2021.106565.

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